

# REPORT DOCUMENTATION PAGE

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# 1995 ONR Transducer Materials and Transducers Workshop

## FINAL AGENDA

4-6 April 1995

### Monday, 3 April 1995

7:00-9:00 p.m. Registration – Scanticon Conference Center

### Tuesday, 4 April 1995 (Second Floor – Room "R")

7:30-8:00 a.m. Registration

8:00-8:15 a.m. A Navy Perspective on Transducer Materials and Device  
Kenneth Dial (Office of Naval Research)

8:20-8:50 a.m. Navy Needs in Transduction Materials  
E.F. Rynne (NCCOSC, San Diego)

8:55-9:35 a.m. Transducer Studies in IMRL: Overview  
L. Eric Cross (Materials Research Laboratory, Penn State)

9:40-10:20 a.m. –Break–

10:20-10:40 a.m. SonoPanel<sup>TM</sup> 1-3 Piezocomposite Panels for Sonar and Active Surface Control  
R. Gentilman, L. Bowen, D. Fiore, H. Pham, and W. Serwatka  
(Materials Systems Inc.)

10:45-11:05 a.m. Combined Sensor - Actuator Tile for the NRL-ABC Research Platform  
Robert D. Corsaro and Brian Houston (Naval Research Laboratory)

11:10-11:30 a.m. Shape Electrode Transducer for Producing Low Sidelobe Directivity Patterns Using 1-3 Composite Material  
C.W. Allen and W.J. Hughes (Applied Research Laboratory, Penn State)

11:35-11:55 a.m. Processing of Fine-Scale PZT Fiber and Fiber/Polymer Composites  
A. Safari, V.F. Janas, R.P. Schaeffer, B. Jadidian, and R.K. Panda (Dept. Ceramic Engineering & Center for Ceramic Research, Rutgers University)

12:00 noon *Conferee Lunch – location to be announced*

1:30-1:50 p.m. A Magnetostrictive/Ceramic Hybrid Transducer  
W.J. Hughes (Applied Research Laboratory, Penn State) and  
J.L. Butler (Image Acoustics, Inc.)

1:55-2:15 p.m. High-Power Terfenol-D Flextensional Transducer  
Mark B. Moffett (Naval Undersea Warfare Center, New London); Raymond Porzio and  
Gerald L. Bernier (Antisubmarine Warfare Directorate, Lockheed Sanders, Inc.)

2:20-2:40 p.m. Drive Voltage Dependence of Electromechanical Coupling in Piezoelectric Ceramics  
K. Uchino, S. Takahashi, S. Hirose, and J.H. Zheng (Materials Research Laboratory, Penn State)

2:45-3:05 p.m. Quasi-Static Coupling Coefficients for PMN-X Materials for Sonar Transducers  
S.R. Winzer, M. Massuda, K. Bridger, and S.A. Brown (Martin Marietta Lab)

3:10-3:50 p.m. –Break–

3:55-4:42 p.m. Three Minute Poster Summaries

1. 3-3 PZT/Epoxy Composite Hydrophone from Distorted Reticulated Ceramics  
W.A. Schulze, M.J. Creedon, and S. Gopalakrishnan (Alfred University)
2. Production of Distorted 3-3 Hydrophone Composites from Reticulated Ceramics  
D.A. Norris, T.B. Sweeting, L.A. Strom, and J.R. Morris (Hi-Tech Ceramics, Inc.)
3. Development of Fine Continuous PZT Fibers for Fiber/Polymer Composites  
R. Loh, G. Weitz, R. Cass, J. Luke (Advanced Cerametrics Inc.);  
A. Safari, V.F. Janas, B. Jadidian (Rutgers University)
4. Replication Processing of Fine Scale 1-3 Piezocomposites  
D. Mess (Cambridge Microtech Inc.) and A. Safari (Rutgers University)
5. Fabrication of Piezoelectric Ceramic Fibers using Sol-Gel Technology  
R. Meyer, Jr., J. Witham, S. Yoshikawa, and T. Shrout (Materials Research Laboratory, Penn State)
6. PZT MicroRods<sup>TM</sup> for Actuators and Sensors  
M.V. Parish, D.L. Carnahan, and D.L. Ouellette (CeraNova Corporation)
7. Low Side-Lobe PVDF Element Using A Space Tapered Electrode  
Michael F. Janik (Raytheon Electronic Systems Division)
8. Recent Terfenol-D Transducer Developments  
Jan F. Lindberg (Naval Undersea Warfare Center, New London) and  
John L. Butler (Image Acoustics, Inc.)
9. Magnetostrictive Transducer materials for Low Temperatures  
M. Wun-Fogle (U.S. Naval Surface Warfare Center, Silver Spring); A.E. Clark (Clark Associates);  
J.B. Restorff (U.S. Naval Surface Warfare Center, Silver Spring); and J.F. Lindberg (U.S. Naval  
Undersea Warfare Center, New London)
10. Magnetic and Elastic Properties of Terfenol-D Films  
Q. Su, Y. Zheng, Y. Wen (Dept. of Material and Nuclear Engineering, University of Maryland),  
J.P. Teter (Naval Surface Warfare Center, Silver Spring); A. Roytburd and  
M. Wuttig (Dept. of Material and Nuclear Engineering, University of Maryland)
11. Electrostrictive Ceramic Development and Characterization for Transducer Applications  
J.D. Weigner and D.J. Erickson (Lockheed Martin, Syracuse)
12. PMN-Based Electrostrictive Actuators for Applications in Corrosive Environments  
Suresh Viswanathan and Steven M. Pilgrim (New York College of Ceramics, Alfred University)
13. Molecular Modeling of PMN Ceramics  
G.J. Kavarnos and H. Robinson (Naval Undersea Warfare Center, New London)
14. A Correlation Between Weak and High Field Phase Transition Characteristics in  $Pb(Mb_{1/3}Nb_{2/3})O_3$ -  
 $PbTiO_3-(Ba,Sr)TiO_3$  Relaxor Ferroelectrics  
E.R. Seydel, I.K. Lloyd, S.M. Pilgrim (Alfred University); and  
K. Bridger (Martin Marietta Laboratories)
15. Processing and Dielectric Measurements on  $Pb((MgNi)_{1/3}Ta_{2/3})O_3(PMNNiTa)$   
P. Leidinger (Friedrich Alexander Universität Erlangen Nürnberg); and S.M. Pilgrim  
(New York State College of Ceramics, Alfred University)

Wednesday, 5 April 1995 (continued)

2:45-3:05 p.m. CERAMBOWS: Prestressed Composite Ceramic Actuators  
G.H. Haertling (Department of Ceramic Engineering, Clemson University)

3:10-3:50 p.m. -Break-

3:50-4:29 p.m. *Three Minute Poster Summaries*

17. Multi-Beam, Acoustic Lens Imaging Sonar  
E.O. Belcher (Applied Physics Laboratory, University Washington), L.L. Harman (Echo Ultrasound), B. Johnson (Naval Explosive Ordnance Disposal Technology Division, Indian Lake)

18. Piezoelectric Composite Transducer Arrays for Underwater Imaging Systems  
Charles S. Desilets (UltraSound Solutions)

19. Micromachined PZT High Frequency Sonar Transducers  
J. Berstein, K. Houston, L. Niles (C.S. Draper Laboratories); K. Udayakumar, H. Chen, and L.E. Cross (Materials Research Laboratory, Penn State)

20. Experimental Studies of Piezocomposites. Part I. Comparison with Theoretical Models  
J.R. Yuan, K. Liang, P. Marsh, H.A. Kunkel (Echo Ultrasound); Q.M. Zhang, X.C. Geng, W.W Cao, W.K. Qi (Materials Research Laboratory, Penn State)

21. Arbitrary Linear, Planar and Spatial Acoustic Array Design and Synthesis: Computer Aided Directivity, Instant Visualization and Experimental Verification  
Dehua Huang and Stephen G. Boucher (Airmar Technology Corp.)

22. Design and Fabrication of a Flexi-Distortional Piezoelectric Sensor  
P.G. Chalk, W.B. Carlson, W.A. Schulze, S.M. Pilgrim (School of Ceramic Engineering and Science, New York State College of Ceramics)

23. Antiferroelectric Phase-Switching Thin Films  
S. Trolier-McKinstry, K. Yamakawa, and I.W. Kim (Materials Research Laboratory, Penn State)

24. Kinetics of Hydrothermal Lead Titanate Powder Formation  
B.L. Gersten, M.M. Lencka, and R.E. Riman (Department of Ceramics, Rutgers University)

25. Plasma-Sprayed Lead Zirconate Titanate - Glass Composites  
S. Sherrit, C.R. Savin, H.D. Wiederick, and B.K. Mukherjee (Dept. of Physics, Royal Military College of Canada)

26. The Effect of Chemical Processing Variables on Powder Characteristics of Hydrothermal PZT Powder  
L. Peters, M.M. Lencka, A. Safari, and R.E. Riman (Department of Ceramics, Rutgers University)

27. Origin of Large Pores in Tape Cast PZT  
S.A. Lish and W.R. Cannon (Center for Ceramic Research, Rutgers University)

28. Process and Properties of Lead Titanate for 0-3 Composites  
Manfred Kahn (Naval Research Laboratory)

29. Interaction of AG/PD Metallization with Lead Oxide, and Lead-Based Electroceramics  
W. Huebner (University of Missouri-Rolla) and S.F. Wang (Vitramon Inc.)

30. Electro-Mechanical-Thermal Finite Element Modeling Methodology for Transducer Arrays  
G. Wojcik, D. Vaughan, N. Abboud, J. Mould, Jr. (Weidlinger Associates) (*No Summary*)

4:29 p.m. Poster Session – Room "P"

Wednesday, 5 April 1995 (continued)

Scanticon Conference Center – President's I & II

6:30 p.m. Cash Bar

7:00 p.m. Dinner

After Dinner Talk – "Old Dogs Learn New Tricks in Europe and Asia"

*R.E. Newnham and L.E. Cross*

Thursday, 6 April 1995 (Second Floor – Room "R")

7:30-8:00 a.m. Coffee

8:00-8:20 a.m. Growth and Characterization of Relaxor Ferroelectric Single Crystals

Thomas R. Shrout, George Risch, Maureen Mulvihill, Seung-Eek Park

(Materials Research Laboratory, Penn State); and Zuang Li (Materials Science Division, Argonne National Laboratory)

8:25-8:45 a.m. Advances in the Modeling and Manufacturing of Transversely Aligned Piezoelectric Fiber Composites

N.W. Hagood, A.A. Bent, and J.P. Rodgers (MIT)

8:50-9:10 a.m. Electric Field Forces as a Means to Develop Processing of New Materials

C.A. Randall (Materials Science and Engineering, Penn State)

9:15-9:35 a.m. Moonies, Cymbals, and BBs

R.E. Newnham, A. Dogan, J.T. Fielding, J.F. Fernandez, D. Smith, J. Tressler, J. Wallis, K. Uchino, and W. Zhu (Materials Research Laboratory, Penn State)

9:40-10:20 a.m. –Break–

10:20-11:08 a.m. *Three Minute Poster Summaries*

31. Active Control of Sound with Foam-PVDF Composite Transducers  
C.A. Gentry and C.R. Fuller (Virginia Polytechnic Institute and State University)

32. Composite Smart Materials for Defense and Dual-Use Applications  
S.R. Winzer (Martin Marietta Laboratories)

33. Evaluation of Piezoelectric Composite as Sensor/Actuator Combination in Vibration Control  
J.P. Dougherty and Y. Chen (Materials Research Laboratory, Penn State)

34. A Concurrently Engineered Adaptive Composite Panel  
G.H. Koopman, K.L. Koudela, and W. Chen (Center for Acoustics and Vibration, Penn State)

35. Ceramic-Metal Composite Transducers for Hydrophone Applications  
J.F. Tressler, K. Uchino, and R.E. Newnham (Materials Research Laboratory, Penn State)

36. The Effect of Design on the Characteristics of the "Moonie and Cymbal" Actuators  
A. Dogan, J. Fernandez, K. Uchino, and R.E. Newnham (Materials Research Laboratory, Penn State)

37. The Effect of Materials on the Performance of the "Cymbal" Actuators  
J. Fernandez, A. Dogan, J.T. Fielding, K. Uchino, and R.E. Newnham (Materials Research Laboratory, Penn State)

38. Piezoelectric Composite Materials for Sensor/Actuator Combinations  
Y. Chen and J.P. Dougherty (Materials Research Laboratory, Penn State)

Thursday, 6 April 1995 (continued)

10:20-11:08 a.m. *Three Minute Poster Summaries (continued)*

39. Vibration Isolation Using Piezoelectric Transducers  
V. Hugo Schmidt, George F. Tuthill, Steven C. Meschia, R. Jay Conant, and Angela K. Prien  
(Montana State University)
40. Development of Piezoelectric Ceramics For, and Mechanical Modeling of Traveling Wave Motors  
W. Huebner, D. Stutts, J. Cummings (University of Missouri-Rolla); and C. Mentesana (Allied Signal, Kansas City Division)
41. High Precision Piezoelectric Linear Motors of Operations at Cryogenic Temperatures and Vacuum  
D. Wong, G. Carman, M. Stam (UCLA); Y. Bar-Cohen, A. Sen, P. Henry, G. Bearman, and J. Moacanin (JPL)
42. Acoustic NDE: 1. Residual Stress Measurements in Plastics, 2. Non-Invasive Cure Monitoring of Composites  
Nisar Shaikh (Analytic Engineering Company)
43. The Ferroelectric Behavior and Piezoelectric Properties of Nylon 5,7 Copolymers  
B. Mei, J.I. Scheinbeim, B.A. Newman (Polymer Electroporessing Laboratory, Rutgers University); M. Berlin, and M.H. Litt (Case Western Reserve University)
44. Electrostatics at Rough Interfaces  
D.J. Klingenberg (Department of Chemical Engineering & Rheology Res. Center, Univ. of Wisconsin); and S.L. Cooper (Dept. of Chem. Engr., University of Delaware)
45. The Compliance Coefficients of an AMP (Formerly Pennwalt) PVDF Copolymer  
Alan O. Sykes (Acoustical Research & Applications)
46. Dual Beam Vibrometry Measurements of Electroactive Polymer Films  
E. Balizer (NSWC White Oak Lab); F. Guillot, J. Jarzynski (Dept. of Mech. Engr., Georgia Inst. Tech.)

11:08 a.m. Poster Session - Room "P"

12:00 noon *Conferee Lunch - location to be announced*

1:30-1:50 p.m. Photostrictive Effect in PLZT Ceramics and its Applications  
Kenji Uchino and Sheng-Yuan Chu (Materials Research Laboratory, Penn State)

1:55-2:15 p.m. Field-Induced Strain and Polarization Switching Mechanisms in PLZT Ceramics Near the PZT Morphotropic Phase Boundary  
Xunhu Dai, Z Xu, Jie-Fang Li, and Dwight Viehland (Department Materials Science and Engineering & The Materials Research Laboratory, University of Illinois)

2:20-2:40 p.m. Transverse Piezoelectric Mode Piezoceramic Polymer Composites with High Hydrostatic Piezoelectric Responses  
Q.M. Zhang, H. Wang, J. Fielding, R.E. Newnham, and L.E. Cross (Materials Research Laboratory, Penn State)

2:45-3:25 p.m. -Break-

3:25-3:45 p.m. Homogenization Theory for Piezocomposites and Applications to Acoustic Transducers  
L. Berlyand (Dept. of Mathematics & Mat. Res. Lab., Penn State)

3:50-4:10 p.m. Computer Simulation of Domain Structures and Their Kinetics of Ferroelectrics  
Wenwu Cao (Materials Research Laboratory, Penn State)

Thursday, 6 April 1995 (continued)

4:15-4:35 p.m. Size Effects in Ferroic Solids  
R.E. Newnham (Materials Research Laboratory, Penn State)

4:40 p.m. Adjourn

# PENNSTATE



**Dr. L. Eric Cross**  
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18 September 1997

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Dear DTIC,

Enclosed is the one copy of the *final report* for Grant Number N00014-95-1-0969 titled "ONR Transducer Materials and Transducers Workshop" for the period of 15 April 1995 through 14 April 1997 that we are required to submit to you.

If you need further information please notify me.

Yours sincerely,

L. Eric Cross  
Evan Pugh Professor  
of Electrical Engineering

LEC:tmc  
Enclosures (1)

cc: S. Kunkle (Grants and Contracts)

**1995 ONR Transducer Materials and Transducer Workshop  
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# **1995 ONR Transducer Materials and Transducers Workshop**

## ***Abstract Booklet***

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***ABSTRACTS***

## NAVY NEEDS IN TRANSDUCTION MATERIALS

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Active transducer materials, as exemplified by the lead zirconate titanates, have been critical to the development of reliable, high power sonar. Navy applications, which include both shipborne and deployed sensors, extend over roughly 5 orders of magnitude in frequency; from low frequency, long range systems to high resolution navigation sonars and include both shipborne and deployed sensors. With the emphasis on "right-sizing", there is a need for development of more flexible forces. This increases the interest in smaller, more versatile sonars and therefore in higher energy density active materials. Ongoing work under ONR 6.2 sponsorship is investigating the application of two ceramic materials: magnesium niobate (PMN), an electrostrictor/relaxor, and lead lanthanum zirconate titanate stannate (PLZTS), a material which undergoes a field-induced change from the antiferroelectric to the ferroelectric phase. The relative energy densities demonstrated in these materials suggest potential increases of 7 to 17 dB over Navy Type III piezoceramics. A study of the characteristics of several PMN compositions was recently completed. The characteristics of the PLZTS compositions is more complex due to its nonlinearity and its performance under mechanical load will be measured shortly. In addition to the current work, this paper will discuss Navy needs for both active and structural materials for transducers.

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## TRANSDUCER STUDIES IN IMRL: OVERVIEW

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In materials studies, three areas have been very active: The search for alternative perovskite structure solid solutions which encompass morphotropic ferroelectric:ferroelectric phase transitions is now focused on lead indium niobate:lead scandium tantalate (PIN:PST) and lead scandium niobate lead titanate (PSN:PT). Single crystal as well as ceramic compositions in these systems are of interest.

For the lead zirconate titanate (PZT) system work is continuing on a range of problems related to the domain structure, domain continuity across grain boundaries, and the simulation of structure in constrained grains. Scale effects as a function of grain size are of both theoretical and practical importance. Since many PZT elements are now used as "patches" to control structural vibration through  $d_{31}$  driven response it has become important to explore the behavior of both soft and hard materials as a function of driving and constraint levels.

Interest in phase switching high strain systems has increased markedly with work on multilayer structures in the lead zirconate stannate system (PSnZT). Recently we have demonstrated excellent "square loop" switching in thin film PZSnT compositions which show unusually high energy density and good control at voltages down to 1 volt. A topic of strong interest in these low temperature prepared films is the effect of very fine grain size on these switching compositions.

A very wide range of strain amplifier structures are now under study. New cap designs for the flexextensional (moonie) design show excellent characteristics and by new material selection it has been possible to completely eliminate temperature drift. Very flat response over wide temperature ranges is now possible. For the exciting new rainbow monomorph structures, detailed characterization of the resonant frequency response for a wide range of dimensions has provided a surprisingly simple engineering model for predicting performance.

In composite materials work is continuing on the high loading 0:3 composites for their tractable simplicity. Both tubular 1:3 composites and new designs of 2:2 structure show excellent low frequency/high sensitivity performance. Work is also just commencing on an intriguing new double amplifier concept which combines monomorph and flexextensional concepts.

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## SONOPANEL™ 1-3 PIEZOCOMPOSITE PANELS FOR SONAR AND ACTIVE SURFACE CONTROL

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More than thirty 0.25 x 0.25 meter 1-3 piezocomposite SonoPanel™ transducers were produced and evaluated under an ONR manufacturing demonstration program. The 1-3 composite consists of 15 volume percent PZT-5H rods 1.1 mm diameter x 6.3 mm long in a matrix of soft polyurethane. The polyurethane matrix contains polymer microspheres to tailor the matrix properties. Stiff face plates are then bonded to the 1-3 composite sheet for stress amplification when used as a hydrophone and to enhance the surface response uniformity when used as an actuator. The SonoPanel™ fabrication process has been successfully scaled up for low volume manufacturing.

SonoPanel™ transducer have been tested underwater by NRL and show high RVS and TVR as well as symmetrical beam patterns. Some initial high pressure performance results will be presented.

Next generation SonoPanel™ transducers, with materials and designs optimized for Navy systems are currently being developed as part of a new ARPA smart materials program. Potentially more robust panel designs as well as utilization of PZT-4 ceramic rods in the composite are being investigated. This program is also exploring incorporation of actuators, pressure sensors, and velocity sensors – all utilizing 1-3 composite materials – along with appropriate control electronics into an autonomous smart panel.

This work is supported by the Office of Naval Research and the Advanced Research Projects Agency.

**COMBINED SENSOR - ACTUATOR TILE  
FOR THE NRL-ABC RESEARCH PLATFORM**

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A new research platform has been constructed for underwater structural-acoustics studies of sensor / actuator coupling mechanisms. It consists of a 15 tile array of "ABC" tiles, where the tiles are arranged in a 5 x 3 pattern. These tiles are adhered to a backing structure, which is an air-backed 0.5 inch thick steel plate.

Tiles are 10 inches square, and each tile contains a large area actuator, pressure sensor, and velocity sensor. The actuator material selected is the 1-3 composite, recently available from Material Systems Inc. Using Laser Doppler Vibrometry measurements, we find good uniformity over the 10 inch square surface of the actuator. From freefield TVR measurements, this actuator is found to have an effective piezoelectric constant on the order of  $400 \times 10^{-12}$  C/N, in good agreement with predictions. The large area pressure sensor (hydrophone) uses a nominally 3 mm thick piezorubber layer (PR-307 from NTK Corp.) as the transducer material. The measured freefield sensitivity of this sensor is -193 dB re: V/ $\mu$ pa. The velocity sensor for each tile is a four-element array of Wilcoxon Model 759 accelerometers, where the output signals are summed and integrating (using a custom NRL designed on-board module) to obtain a signal proportional to velocity. These accelerometers have a high resonance frequency (25 kHz) and very low noise floor (approximately 0.10  $\mu$ g/ $\sqrt{Hz}$ ).

This paper presents details of the tile design and the predictive models used. Issues addressed include spatial sampling, nearfield sensing, internal resonances, and both direct and extraneous coupling mechanisms, all of which can contribute to complicate the system transfer functions. Acoustic characteristics of the ABC tile were evaluated in the NRL Large Pool Facility, both in the freefield and when mounted on a backing structure, and these results are compared with predictions. Implications for local control of the actuator's surface are also discussed.

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## **SHAPED ELECTRODE TRANSDUCER FOR PRODUCING LOW SIDELOBE DIRECTIVITY PATTERNS USING 1-3 COMPOSITE MATERIAL**

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Previous studies by Hughes and Allen<sup>1</sup> have shown that shaped hydrophones using PVDF piezoelectric material produce directivity patterns with low sidelobes without the use of multiple elements or shading electronics. In order to obtain similar performance for transmitting sound, 1-3 composite piezoelectric material transducers with shaped electrodes have been fabricated and tested. The transducer uses Material Systems, Inc. 1-3 composite material (30% volume, voided matrix) since it exhibits excellent lateral decoupling between the PZT rods. Measurements show that the shaped transducer produces low sidelobes over a broad frequency range (25 to 200 kHz) and the beamwidths agree well with theory. Sidelobes are as low as -32 dB at 100 kHz, which compares well to the theoretical design value of -35 dB.

<sup>1</sup>Hughes, W. J. and Allen, C. W., "A Shaped PVDF Hydrophone for Producing Low Sidelobe Beampatterns", 1992 Symposium on Autonomous Underwater Vehicle Technology, 2-3 June 1992.

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## PROCESSING OF FINE-SCALE PZT FIBER AND FIBER/POLYMER COMPOSITES

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The processing of fine-scale PZT fiber and fiber/polymer composites for transducer applications is discussed. Emphasis is placed on inexpensive techniques that can form composites with 1-3 connectivity and be easily utilized by composites manufacturers. Progress in several areas, including processing of PZT fibers, development of piezoelectric composites using PZT fibers and tapes, and formation of composites via a modified lost mold method and the replication process, will be reported. (1) PZT fibers: fibers, 10 to 30  $\mu\text{m}$  in diameter, were formed at Advanced Cerametrics, Inc. using the Viscous Suspension Spinning Process (VSSP). Yarns containing between 10 and 500 individual fibers were collimated by applying a polymeric sizing to the yarns, and passing the yarns through sizing dies. Fiber density in the fired yarn were controlled by binder system and die size. These yarns will be woven in different architectures to create composites with novel microstructures. (2) Fiber/polymer composites: above mentioned sized PZT yarns were bundled, fired, and backfilled with polymer to create composites with 1-3 connectivity. (3) Tape laying method: stacked PZT tapes, as fine as 20- $\mu\text{m}$  thick, were backfilled with polymer to make composites with 2-2 connectivity. When diced, these structures yielded fine 1-3 piezoelectric ceramic/polymer composites. Composites with a PZT volume fraction gradient have been demonstrated. (4) Modified lost mold process: the procedure for manufacturing the sacrificial plastic mold was modified to allow rapid prototyping of 1-3 composites with novel spatial scale and periodicity. (5) Replication process: activated carbon fabrics of various architectures, composed of individual 10-20  $\mu\text{m}$  fibers, were soaked in a PZT precursor solution before arrangement into desired structures. The structures were fired, yielding a ceramic relic with the same fiber arrangement as the carbon fiber template. Relics were embedded in an epoxy matrix, polished, electroded and corona poled.

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## **A MAGNETOSTRICTIVE/CERAMIC HYBRID TRANSDUCER**

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A hybrid magnetostrictive/ceramic tonpilz transducer uniquely combines the characteristics of ceramic and magnetostrictive material properties to make a tonpilz transducer with different but desirable characteristics. In the original hybrid design, the three mass system was optimized to attain a high front to back ratio by adjusting the waves to cancel at the tail mass and add at the front mass. It successfully obtained back cancellation in a small physical envelope. The hybrid transducer has also been shown to exhibit wide bandwidth performance by increasing the mass of the central and tail masses. Because of the opposite nature of the piezoelectric and magnetostrictive materials: the transducer is self tuned giving a reduced low frequency response; and has a 90° phase shift between the two sections resulting in a smooth transition between the two resonance frequencies of the system. These two effects yield a nearly uniform response over a frequency band greater than one octave.

## HIGH-POWER TERFENOL-D FLEXTENSIONAL TRANSDUCER

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A flextensional underwater acoustic projector, powered by Terfenol-D and operated at a depth of 400 ft (122 m), has been driven to a source level of 212 dB// $\mu$ Pa-m at NUWC's Seneca Lake Test Facility. The flextensional shell was a modified Class IV shape having concave, rather than convex, radiating surfaces. The geometry of the shell was similar to one described by Merchant [U. S. Patent No. 3,258,738, June 28, 1966]. The shell cross-section was circumscribed by a rectangular area 9.8 x 5.0 inches (0.250 m x 0.126 m), and the height between the end flanges was 5.7 inches (0.145 m). At the resonance frequency of 930 Hz, the projector was omnidirectional, and so the radiated acoustic power was 14 kW. The ac efficiency was 52 percent, and the overall (ac + dc) efficiency was 45 percent. The mass of the transducer was 34 lbm (15.4 kg). With a mechanical quality factor,  $Q_m$ , of 4.7, the projector figure-of-merit exceeded 200 W/kg-kHz-Q. The effective coupling factor, measured with the transducer in air, was 0.29. [Work sponsored by the Office of Naval Research]

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## DRIVE VOLTAGE DEPENDENCE OF ELECTROMECHANICAL COUPLING IN PIEZOELECTRIC CERAMICS

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High power piezoelectric devices such as ultrasonic motors are now intensively developed, where piezo-actuators are driven under rather high applied voltages. The catalogue data for the electromechanical coupling indicated by their manufacturers are usually determined under a low applied voltage (1 V) with an impedance analyzer and sometimes disagree with the high voltage experiments.

This paper describes three new measuring techniques for high-voltage piezoelectric constants, which have been recently developed by us; piezoelectric resonance methods with a constant voltage circuit and with a constant current circuit, and a pulse drive method. The former two methods provide heat generation during the measurement. On the contrary, the latter is not associated with the temperature rise. The electric field dependence of the piezoelectric constant, permittivity, elastic compliance and electromechanical coupling factor was obtained for the same  $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$  based sample with these techniques, and the results were compared each other.

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# QUASI-STATIC COUPLING COEFFICIENTS FOR PMN-X MATERIALS FOR SONAR TRANSDUCERS

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The coupling coefficient of an electro-active ceramic is an important measure of the material's ability to transform electrical energy into mechanical work. The efficiency of the material in converting electrical energy into mechanical energy greatly influences the design of the sonar transmit system, including power amplifiers, cable sizes, winches and other elements, and has a considerable influence on the bandwidth capability. Measurement and interpretation of coupling coefficients in piezoelectric materials are well established. The same cannot be said for electrostrictive ceramics. Similar to all other properties of relaxor ferroelectrics, they are influenced by the electric field, temperature and frequency. Determination of the quasi-static coupling coefficient requires measurement of the electrostrictive coefficients,  $Q_{11}$  and  $Q_{12}$ , the saturation polarization ( $P_s$ ), the materials constant ( $k$ ) and the elastic modulus ( $Y$ ). Based on constitutive models developed by Hom and Shankar (1994) and Hom et al (1994), we have developed measurement techniques and calculated quasi-static coupling coefficients for several compositions of PMN-X at prestresses up to 10,000 psi and temperatures between -20°C and + 60°C. For the PMN-X compositions measured thus far, the highest coupling coefficient is 0.55 at 5°C and 0-3000 psi prestress. Some compositions showed degradation of the coupling coefficient under prestress, while the majority were essentially unchanged. These results, along with their effects on sonar performance will be reported.

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**4 April 1995**

***Poster Summaries***

### 3-3 PZT/EPOXY COMPOSITE HYDROPHONES FROM DISTORTED RETICULATED CERAMICS

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Lead zirconate-titanate (PZT)/epoxy composite hydrophones with 3-3 connectivity have been fabricated by embedding reticulated PZT ceramics in an epoxy matrix. The hydrostatic properties of these composites were improved by distorting the ceramic structure from a nearly isotropic network of PZT ligaments toward a highly oriented, laterally reinforced 1-3 configuration (See Table 1). The composites fabricated with Spurr epoxy exhibit good pressure stability and moderate hydrostatic properties (Fig. 1). Investigation of ways to improve the properties further while maintaining manufacturability are ongoing. Increased distortion of the structure and selection of a more compliant matrix are two of the areas being actively studied. This type of composite also allows consideration of stiff electrode configurations without significant increases in fabrication complexity.

(This work is sponsored by the Office of Naval Research, Grant No. N00014 -92-J-4025)

Table 1. Properties of Reticulated Ceramic Composites with Spurr Epoxy Matrix

Distortion Aspect Ratio	Density (g/cm <sup>3</sup> )	K	d <sub>33</sub> (pC/N)	d <sub>h</sub> (pC/N)	g <sub>h</sub> (x 10 <sup>3</sup> V-m/N)	d <sub>hgh</sub> (x 10 <sup>-15</sup> m <sup>2</sup> /N)
1:1	1.75	96	88	19	23	437
2:1	1.90	143	152	30	24	720
3:1	2.33	268	203	50	21	1050

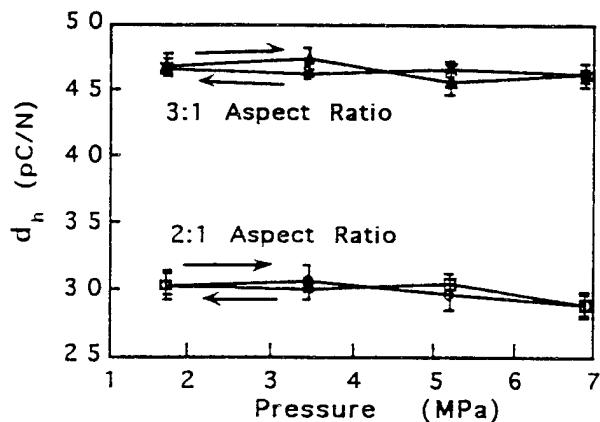


Figure 1. Pressure stability of the hydrostatic charge coefficient of reticulated ceramic composites with Spurr epoxy matrix.

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## PRODUCTION OF DISTORTED 3-3 HYDROPHONE COMPOSITES FROM RETICULATED CERAMICS

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The manufacturing aspects of fabricating 3-3 PZT/epoxy composite hydrophones from distorted reticulated ceramics are being developed in conjunction with research being done on these composites at the New York State College of Ceramics, at Alfred University. Equipment for distorting the polyurethane foam precursor for the ceramic phase of the 3-3 composite has been built, and distorted foams of predictable aspect ratio up to 5:1 can be reproducibility made. The design for this equipment resulted from initial work done by the NYSCC on 3-3 composite hydrophones based on reticulated ceramics technology. The reticulate ceramic process begins with an open cell organic foam which is shaped, dipped into a ceramic slurry and fired to remove the organic and sinter the ceramic. Researchers at the NYSCC reported at the 1994 ONR Review Meeting that PZT distorted foam reticulated ceramics incorporated into a 3-3 hydrophone composite had enhanced hydrostatic response over undistorted foam and showed promise as low density hydrophones. The manufacturing concerns of a consistent distorted foam precursor, a uniform web structure and consistent density among multiple samples are being addressed by Hi-Tech Ceramics. PZT/epoxy composites have been fabricated at 15 to 20 volume percent ceramic from 15, 30, 45 and 65 pore per inch (ppi) reticulate. These pore sizes range from approximately 4 to 1 mm respectively before firing. The greatest  $d_{hgh}$  (figure of merit) obtained has been 1000. An increased response may be possible by optimizing the epoxy phase with fillers or by using an alternate epoxy to the ones tried to date. This area is currently be researched.

DEVELOPMENT OF FINE CONTINUOUS PZT FIBER  
FOR FIBER/POLYMER COMPOSITES

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This is a one year experimental research program focused on the development of fine PZT fibers and on the development of fabrication methods for manufacturing large area piezoelectric composites. The process of wet spinning continuous PZT fiber by powder-in-viscose process is discussed. Emphasis is placed on the precursor preparation, wet spinning process which results in PZT green fiber with 36 microns in diameter and 100 feet in length, and the fiber sintering process. The green fiber solids loading during spinning process and the fiber diameters in different process stages are also discussed. Fiber sizing process and parameters which influence the sized fiber strength and flexibility are reported.

## REPLICATION PROCESSING OF FINE-SCALE, 1-3 PIEZOCOMPOSITES

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Composites of piezoelectric ceramics and polymers have shown promise as advanced transducer materials. An ideal material would have good electromechanical coupling, a large hydrophone figure of merit, and an acoustic impedance close to that of water.

We are evaluating a method of forming fine scale composites with 1-3 connectivity. In this work, we extend the replication processing technique to a fine scale (ceramic fiber diameter less than 20 microns) by employing a suitable template material. Microtubular plastic fibers are excellent candidate materials for the formation of composites with approximately 10 volume percent ceramic phase.

Hollow polyester fibers (outer diameter 30 microns, inner diameter 10 microns) have been mounted into a collimated bundle. The ends of the bundle are sealed with a urethane rubber, and then microtomed so that the fiber interiors but not the interfiber voids are accessible. The bundle is then vacuum infiltrated with a sol-gel PZT feed, or alternately a zirconia sol. The liquid-filled bundle is then exposed to ammonia gas, causing an in situ gelation, which yields an array of parallel, non-intersecting ceramic gel fibers. In a heat treatment step, the polyester and urethane is burnt off to yield a large number of very high aspect ratio ceramic fibrils. Fibril diameter is approximately 6 microns, and length is over 200 microns. However in undergoing the gel-to-oxide transition, we are experiencing difficulty in retaining the aligned geometry of the fibril array. Work in progress is directed toward forming a more stable fibril array of somewhat lower aspect ratio.

## FABRICATION OF PIEZOELECTRIC CERAMIC FIBERS USING SOL-GEL TECHNOLOGY

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An investigation was carried out on the preparation of ultra-fine scale ( $<100\mu\text{m}$ ) lead zirconate titanate (PZT) and barium strontium titanate (BST) fibers derived from metal alkoxide precursors. A systematic study of variables that influence fiber drawing and fiber properties was conducted to determine the optimum processing conditions necessary for fabricating high quality fibers. Variables such as curing temperature, hydrolysis ratio, acid type and concentration, and firing temperature were investigated to link processing conditions to fiber drawing ability and final fiber properties. PZT fibers and gels were characterized by x-ray diffraction, thermal gravimetric analysis, differential thermal analysis and scanning electron microscopy. Preliminary electrical characterization methods and electrical properties will be discussed along with potential applications for fiber composites.

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## PZT MicroRods™ FOR ACTUATORS AND SENSORS

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Fine uniform diameter barium titanate ( $\text{BaTiO}_3$ ) and several types of lead zirconate titanate (PZT) piezoelectric MicroRods have been fabricated in semi-continuous lengths using an extrusion method. A heat treatment procedure has been developed to sinter the filaments to near-full density resulting in controlled diameters that range from 100  $\mu\text{m}$  or less to 1 mm with lengths from a typical 10 cm to semi-continuous (meters). Diameter standard deviations of the finest filaments have been reduced from less than 5% to around 2%. Our process is being scaled from the production of kilometers to tens-of-kilometers for economical use in sensor and actuator composites. A method of poling piezoelectric filaments is being developed that will allow continuous poling in axial or transverse directions. In addition, a comparative method using a force gauge signal was developed to yield  $g_{33}$  constants.

CeraNova will report on the latest electrical, mechanical, and processing results. In addition, CeraNova is currently using their piezoelectric MicroRods in the development of sensor composites. Individual PZT MicroRods have been poled under a number of times, temperatures and fields, followed by a measurement of forces produced relative to a standard while applying a voltage. Preliminary electrical property measurements show the  $g_{33}$  constant of these filaments to be equivalent to the powder manufacturer's published values for bulk samples ( $\sim 600 \times 10^{-12} \text{ Vm/N}$ ). Preliminary mechanical property measurements show tensile strengths equivalent to those of other bulk specimens, though a much more detailed analysis is underway.

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## Low Side-Lobe PVDF Element Using A Space Tapered Electrode

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This paper describes an approach to hydrophone element design which results in a beam pattern with a low side-lobe structure. The method used is called space tapering. Space tapering simulates a conventional amplitude taper (e.g. Dolph-Chebyshev or Taylor) by varying the spacing of equally sized segments of electrode material. These segments are tied in parallel to produce the element output. This method is effective for single element beam configurations. Single element beams with space tapered electrodes provide improved spatial filtering. It combats the problem of unwanted reverberation and ambient noise entering through the sidelobe structure and lowering the resulting SNR. Spatially shaded elements are also effective in array designs. Since the beam output is the multiplication of the element response and the beam response of an array of point sources, changing the element spatial response is an alternative effective method of shading the array. Spatially shaded elements can be used with or without additional array amplitude weighting. This paper will discuss the design, construction, and test of five prototype elements which were fabricated on a single sheet of Polyvinylidene Difluoride (PVDF). The five elements consisted of two control elements and three elements with increasing degrees of spatial tapering. The results show that, as the spatial density of the segments increase, the beam pattern approaches the ideal theoretical prediction.

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## RECENT TERFENOL-D TRANSDUCER DEVELOPMENTS

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Terfenol-D, a magnetostrictive rare earth alloy of terbium, dysprosium, and iron, has recently been utilized in the design of two different transduction devices with striking results.

The Class VII flexensional transducer, patented by Merchant in 1966 and referred to as the dogbone or peanut shell flexensional because of its shape, was never reduced to practice until quite recently when Ray Porzio designed and built a dogbone flexensional with Terfenol-D drive as part of a project at NUWC. It has been found that the low sound speed of the magnetostrictive material combined with the high compliance of the dogbone shell enhances the inherent bandwidth of the transducer. In a separate paper at this conference Moffett et al. detail the exemplary performance of a DC-biased dogbone flexensional which exhibited a transducer figure-of-merit of 200 (approximately seven times greater than a traditional Class IV flexensional with PZT drive). This paper will discuss a Terfenol-D dogbone flexensional with permanent magnet bias designed and built in parallel with the transducer being reported by Moffett. The design, which utilized neodymium-boron-iron magnets interspersed in the Terfenol-D drive rods and a tapered shell, achieved a figure-of-merit of 113 before experiencing heating problems which affected the magnet performance.

A magnetostrictive/piezoelectric hybrid tonpilz transducer was patented in 1984 by Butler and Clark and subsequent development under the auspices of the Navy's Small Business Innovation Research (SBIR) program at NUWC resulted in the design and fabrication of a 12 element array which exhibited excellent acoustic performance (Butler et al., J. Acoust. Soc. Am. 94, pp 636 (1993)). Recent efforts by Butler with manipulation of the design parameters have resulted in analytical results predicting a vastly increased bandwidth of the transducer. An increase in bandwidth from a traditional tonpilz value of 40% to over 100% was predicted. Prototype transducers and a partial array have been fabricated by Dr. Jack Hughes at the Applied Research Laboratory at the Pennsylvania State University and measurements have confirmed the predicted broadband characteristics. [Work sponsored by the Office of Naval Research]

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## MAGNETOSTRICTIVE TRANSDUCER MATERIALS FOR LOW TEMPERATURES

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It has been recently shown that large magnetostrictive strains and magnetomechanical coupling factors are available at low temperatures in materials containing the elements terbium and dysprosium.<sup>1,2</sup> At 77 K strains exceed 0.5% with the application of magnetic fields of a few hundred Oersteds ( $\sim 10$  kA/m). Coupling factors ( $0.6 < k < 0.9$ ) depend upon the magnetic field, compressive stress and temperature. In a 430 Hz transducer application, excitation at  $\sim 77$  K was made via lossless high temperature (BiSrCaCuO) superconducting wire.<sup>3</sup> More conventional (NbTi) superconducting coils are used for applications at lower temperatures. Using a binary alloy of terbium and dysprosium (Tb<sub>0.76</sub>Dy<sub>0.24</sub>), a superfluid leak-tight low temperature valve was constructed for operation below 4 K.<sup>4</sup> Another promising low temperature magnetostrictive compound, TbZn, is machinable and can be bolted directly into transducer structures. This material has a useful temperature range from 4 K to 150 K. A saturation strain of  $\sim 0.5\%$  and coupling factor  $\sim 0.9$  were found at 77 K. Materials tailored with high magnetic anisotropy can convert large amounts of stored internal energy ( $\sim 200$  kJ/m<sup>3</sup>) to work with the application of only small changes of magnetic field. Such materials can be used as magnetically excited switches with large force capabilities. In this presentation, the performance of three high strain magnetostrictive materials will be compared: Tb<sub>x</sub>Dy<sub>1-x</sub> hexagonal alloys, TbZn-based cubic compounds, and the high temperature Tb<sub>x</sub>Dy<sub>1-x</sub>Fe<sub>2</sub> (Terfenol-D) intermetallic compounds.

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## MAGNETIC AND ELASTIC PROPERTIES OF TERFENOL-D FILMS

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Glassy films of Terfenol-D,  $Fe_2(Dy_{0.3}Te_{0.7})_1$ , were routinely deposited under 7 mTorr Ar pressure with no intentional substrate heating by DC magnetron sputtering. The substrates used were Si/SiO<sub>2</sub> cantilevers with 200 nm SiO<sub>2</sub> serving as a diffusion barrier and Si membranes. The deposition rate was 0.2 nm/sec for all films which had the target alloy composition. The magneto-elasticity, anelasticity and ferromagnetic properties of the so prepared films was studied yielding information on the film evolution, domain structure, modulus softening, damping and magnetization characteristics.

The kinetics of the evolution of the Terfenol-D film and domain structure during and immediately after deposition was studied by in-situ measurements of the eigenfrequency of Si membranes onto which Terfenol-D was deposited. The results show that the films are entirely reconstructed after each deposition by a process which is thermally activated.

Atomic Force Microscopy reveals a periodic one-dimensional domain structure in as deposited compressed films. An analysis based on the theory of twinning shows that these characteristics are a consequence of the elastic film/substrate interaction proven by the linear dependence of the square of the domain period as a function of the film thickness. The alignment of this one dimensional structure follows the elastic anisotropy of the substrate providing further support of the importance of this interaction.

The magneto-elastic response of Terfenol-D films is greatest in films under zero stress. At this point the  $\Delta E$  effect of a 2 $\mu$ m Terfenol-D/45 $\mu$ m Si composite equals 20% signalling that the  $\Delta E$  effect of the films is almost 100%, i.e. that the effect is almost totally controlled by domain wall motion. Since the state of stress can be manipulated by thermal treatment, the magneto-elastic properties of Terfenol-D films can be controlled as well.

This study was supported by the Office of Naval Research, contract No. N00014-93-10506. It also benefitted from support of the National Science Foundation, Grant No. DMR-93-21185 and the Army Research Office, contract No. DAAL03-92-G-0121.

ELECTROSTRICITIVE CERAMIC DEVELOPMENT AND  
CHARACTERIZATION FOR TRANSDUCER APPLICATIONS

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During the past year two research projects were performed to investigate electrostrictive ceramics and transducers. The first was the development of fabrication techniques for lead magnesium niobate (PMN) electrostrictive powders for sonar transducer applications. The second project involved fabricating ceramic rings for use in an electrostrictive flex-tensional transducer. A mass loaded balanced resonator was assembled using the PMN rings to obtain fundamental transducer data useful to the transducer designer.

The ceramics development stemmed from work done at the Lockheed Martin Laboratory-Baltimore on compositions developed for Navy use in sonar applications. Two eight-kilogram batches and one 15-kilogram batch were fabricated using the columbite process. Lead and magnesium levels were varied and the materials evaluated for optimum properties. Transverse microstrain in excess of 300 ppm with low hysteresis was obtained with material properties optimized for use at 5°C.

A 22-ring PMN stack (ring size 2.6"O.D.x1.37"I.D.x0.375" thick) was assembled into a heavily mass-loaded resonator assembly and tested in air. Basic material properties such as dielectric constant, coupling factor, elastic modulus, etc. were obtained. The PMN stack was composed of cemented rings and compared to a mechanically identical Navy Type III PZT stack. The reference data obtained from the PZT stack were used to extrapolate room temperature material data for PMN. The values obtained indicate coupling=0.54;  $d_{33} = 967$  pC/N; and dielectric constant= 21,000.

The authors wish to acknowledge the assistance of the Lockheed Martin Laboratory in Baltimore for material analysis and property measurements performed on the various powder batches.

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## **PMN--BASED ELECTROSTRRICTIVE ACTUATORS FOR APPLICATIONS IN CORROSIVE ENVIRONMENTS**

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Ultrasonic investigation forms an important tool for various applications ranging from high frequency medical ultrasound imaging through acoustic nondestructive testing to low frequency sonars. However, it has not yet been applied to high temperature systems as the existing acoustic transducers are limited by their Curie temperatures. These limitations could be overcome with the use of electrostrictive materials; these materials operate above their Curie temperatures and can produce higher mechanical output with low losses and comparable input power.

The reported work explores the feasibility of using Lead Magnesium Niobate (PMN) based acoustic transducers for in-situ monitoring and processing of glass. It involves design and fabrication of PMN--based transducers and evaluating their performance by measuring the thickness of the glass tank refractory block (fused AZS and zircon) at elevated temperatures. Finite element techniques have been used to refine the preliminary design to improve expected performance at elevated temperatures. Pulse-echo measurements at progressively higher temperatures will determine the penetration depth and acoustic attenuation of the refractory. These results will establish the feasibility of using PMN--based acoustic transducers in other similar corrosive environments. At this stage, acoustic evaluation of AZS and fused Zircon have shown that penetration depths of 0.11 m and 0.31 m can be achieved at room temperatures. A more detailed overview of the design and fabrication of the multilayer PMN--based actuator will be presented.

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## MOLECULAR MODELING OF PMN CERAMICS

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Extended Hückel molecular orbital theory was used to analyze the orbital interactions in  $\text{PbNbO}_3^{1+}$  and  $\text{LaNbO}_3^{2+}$  model structures representative of PMN ceramic. These structures were chosen to determine the orbital effects, if any, that an A-site substitutional such as  $\text{Pb}^{2+}$  or  $\text{La}^{3+}$  has on bond stability in the crystal lattice structure. It was determined that the A-site ion does not directly influence bonding between the A-site ion and atoms in the neighboring crystal lattice but does change the position of the Fermi level, which in  $\text{PbNbO}_3^{1+}$  is -10.1 eV and in  $\text{LaNbO}_3^{2+}$  is -14.5 eV. In  $\text{PbNbO}_3^{1+}$ , the Fermi level is so positioned that the bonds linking Nb and O are destabilized. In contrast, there is no antibonding character in  $\text{LaNbO}_3^{2+}$ . The shifting of the Fermi level as a function of the A-site ion is used to rationalize the experimental observation that pure PMN does not coarsen or undergo additional crystallization during annealing, but La-substituted PMN does in fact favor ordering of the crystal structure.

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A CORRELATION BETWEEN WEAK AND HIGH FIELD PHASE  
TRANSITION CHARACTERISTICS IN  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $\text{PbTiO}_3$ -  
 $(\text{Ba},\text{Sr})\text{TiO}_3$  RELAXOR FERROELECTRICS

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The quantity  $Q_{\text{eff}}$ , the effective electrostrictive coefficient, is defined as the induced strain for a given electric field divided by the square of the induced polarization.  $Q_{\text{eff}}$  is useful as a parameter for indicating the transition behavior of  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $\text{PbTiO}_3$  (PMN-PT) based electrostrictors at high electric fields.  $Q_{\text{eff}}$  is also useful for correlating weak and high field behaviors for a number of compositions. Transverse strain and induced polarization were measured simultaneously at various fields for several compositions of PMN-PT doped with  $\text{BaTiO}_3$  or  $\text{SrTiO}_3$ . The transition temperature between the primarily piezoelectric state and the principally electrostrictive state is indicated by changes in the value of  $Q_{\text{eff}}$ .  $Q_{\text{eff}}$  plotted against temperature shows a sudden change in slope consistent with the onset of a field induced ferroelectric phase transition. A temperature scale normalized to the temperature at which each composition shows a maximum in relative permittivity,  $T-T_{\text{max}}$ , displays a good correlation for all the compositions. The field induced phase transition occurred at  $T-T_{\text{max}} = -56^\circ\text{C} \pm 2^\circ\text{C}$  for all four of the tested compositions at 0-1.0 MV/m and 10 Hz. At frequencies of 100 Hz and higher, the effects of hysteretic heating when measuring strain and polarization simultaneously are to create distinct identifiable distortions of the data.

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## PROCESSING AND DIELECTRIC MEASUREMENTS ON $Pb(MgNi)_{1/3}Ta_{2/3}O_3$ (PMNiTa)

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The aim of this work is to investigate the dielectric properties and the electrostrictive effect in the perovskite  $Pb(Mg_{1/3}Ta_{2/3})O_3$  -  $Pb(Ni_{1/3}Ta_{2/3})O_3$ . PMNiTa should have useful electrostrictive properties at low temperatures. The present problem is the formation of undesirable pyrochlore, a crystal structure with low electrostriction and low dielectric characteristics.

To avoid the formation of pyrochlore the columbite precursor route was employed prereacting in a first calcination step  $MgO$ ,  $NiO$  and  $Ta_2O_5$  at a temperature of  $1100^\circ C$  for 6 hours.  $PbO$  was added in a second step. Pellets with between 0 % and 100 % nickel substitution for magnesium were pressed. To minimize lead volatilization the pellets were packed in sacrificial powder in a closed alumina crucible. The firing took place at  $550^\circ C$  for 2 hours followed by a 4 hour hold at  $1250^\circ C$ .

Color differences and XRD showed that several samples had high-perovskite cores and a pyrochlore surface layer, whereas others consisted of pure pyrochlore. As a figure of merit for the perovskite yield the intensity of the 100% peak of the perovskite pattern was divided by the sum of the 100 % peaks for both the perovskite and pyrochlore pattern:

$$\text{Perovskite Yield} = \frac{\text{Maximum Intensity in Perovskite Pattern}}{\text{Sum of Maximum Intensities in Pyrochlore and Perovskite Pattern}}$$

Samples with up to 20 % nickel substitution for magnesium formed at least some perovskite. Those with 20 % substitution showed the highest yield with 94 % perovskite. In samples with 30 % and more nickel substitution for magnesium no perovskite was present.

To carry out dielectric measurements gold electrodes were fired onto the pellets. Relative permittivity  $\kappa'$  and dielectric loss  $D$  were measured at four frequencies (1 kHz, 10 kHz, 100 kHz, and 1 MHz) while decreasing the temperature from  $50^\circ C$  to  $-190^\circ C$  at  $2^\circ C/min$ .

Initial results for the composition with the largest perovskite yield show typical relaxor behavior. For the highest frequency a maximum permittivity of 4700 is reached at  $-100^\circ C$ . Maximum loss is 0.18. For the lowest frequency a maximum permittivity of 5450 is reached at  $-120^\circ C$ . In this case the maximum loss is 0.12.

Results of the permittivity and dielectric loss determinations for the remaining compositions and their relationship to phase content will be presented. These data will be considered in conjunction with electrostrictive characteristics of the materials to assess the usefulness of PMNiTa.

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# STUDIES ON THE ELECTROMECHANICAL PROPERTIES OF PbTiO<sub>3</sub>-BASED CERAMICS

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Lead titanate (PbTiO<sub>3</sub>) exhibits many interesting characteristics including highly anisotropic piezoelectric properties, a high piezoelectric voltage coefficient due to its lower dielectric constant, a high Curie temperature, a high spontaneous polarization, a low aging rate, and a relatively low mechanical quality factor. These attributes make PbTiO<sub>3</sub>-based materials attractive for many bulk, thin film, composite, and single crystal applications.

In our lab we have been performing several studies on modified lead titanate compositions with the purpose of determining whether simultaneous Ca and Sm additions result in desirable piezoelectric and dielectric properties similar or superior to previously-prepared systems. Results showed that the (Pb<sub>0.88-x</sub>Ca<sub>x</sub>Sm<sub>0.08</sub>)(Ti<sub>0.98</sub>Mn<sub>0.02</sub>)O<sub>3</sub>; x = 0.11, 0.13, 0.15 and 0.17 system exhibits normal ferroelectric behavior, with most properties similar to other modified lead titanates. The unique attribute of this system appeared for the x=0.13 and 0.15 compositions, which exhibited k<sub>t</sub>'s  $\geq$ 57%.

In another series of studies the phase diagram of the (Pb<sub>0.85</sub>Sm<sub>0.10</sub>)(Ti<sub>0.98</sub>Mn<sub>0.02</sub>)O<sub>3</sub> - BiFeO<sub>3</sub> system was studied by measuring the dielectric behavior as a function of temperature, coupled with room temperature XRD and measurements of d<sub>33</sub>, k<sub>t</sub>, and k<sub>p</sub>. A solid solution forms over a wide range between the modified PT and the BF, with a mixed phase region occurring between 57.5 and 60%. The maximum K', k<sub>t</sub>, k<sub>p</sub> and d<sub>33</sub> were exhibited in this MPB region. Some of the studied compositions exhibit electromechanical properties suitable for transducer applications, over a compositional range where the K' varies widely.

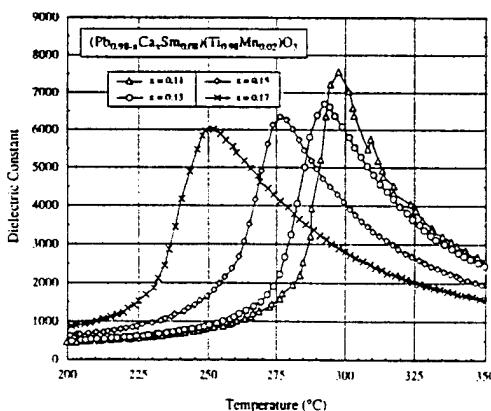


Table I  
Electrical Properties of (Pb<sub>0.88-x</sub>Ca<sub>x</sub>Sm<sub>0.08</sub>)(Ti<sub>0.98</sub>Mn<sub>0.02</sub>)O<sub>3</sub>

	% Ca			
	11	13	15	17
Density (g/cm <sup>3</sup> )	7.19	7.13	7.09	7.04
$\epsilon$ ( $\omega=1$ kHz, $T=25^\circ\text{C}$ )	212	228	251	288
$\tan \delta$ ( $\omega=1$ kHz, $T=25^\circ\text{C}$ )	0.16%	0.10%	0.17%	0.10%
T <sub>c</sub>	298	294	278	251
d <sub>33</sub> (pC/N)	64	67	72	81
k <sub>t</sub> , %	47.6	57.0	57.2	54.8
k <sub>p</sub> , %	=0	=0	=0	=0
Q <sub>m</sub>	82	29	39	29
N <sub>f</sub> (MHz-mm)	2.259	2.168	2.156	2.237

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***ABSTRACTS***

## THE EFFECTIVENESS OF SPARSE RANDOM ARRAYS FOR UNDERWATER ACOUSTIC IMAGING

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The current innovation program underway in Australia to produce an underwater imaging system suitable for turbid waters has highlighted the need to trade off complexity with performance. Sparse arrays reduce the manufacturing complexity to the level of current capability while random elements overcome some of the defects of widely spaced array elements.

Random arrays have been utilised for twenty years in microwave systems, but they have rarely been used in acoustic systems. Yet these sparse arrays have desirable properties; in particular, the resolution achieved is undiminished from that of a filled array of the same aperture. The main drawback is a raised sidelobe level at angular displacements beyond the first few sidelobes. In the continuous-wave case, the average intensity in these distant sidelobes is independent of angular displacement and is equal to  $1/N$  (relative to the peak intensity), where  $N$  is the number of elements. But, for arrays whose aperture is sufficiently large compared to the wavelength, this level can be made quite low while preserving sparseness. Overall, random arrays are believed to be very cost-effective for high-resolution imaging, especially for two-dimensional (as opposed to one-dimensional) arrays.

We have been engaged with others on preliminary design work for an underwater acoustic camera using a random array. The image is to have of order  $1000 \times 1000 \times 1000$  resolution cells or volume pixels, with the target body essentially filling the whole image. The bandwidth of the signal is to be comparable with the central frequency, rather than much smaller. The image is to be of an angular field of order  $90^\circ \times 90^\circ$ .

We have performed computer emulations of image-forming for array systems appropriate to the above design, with emphasis on the image of a point target. Included are near-field focussing using exact path lengths, and also the option of obtaining good range resolution, through either a short toneburst or a cross-correlated chirped signal. It is found that the performance of the random array is undiminished by these inclusions; in fact, both the average level and the peak level of the distant sidelobes are in general improved. Also the performance of partially random arrays, constructed out of identical subarrays for ease of manufacture, has been investigated.

## UNDERSEA ACOUSTIC COHERENCE AT 3-5 MHZ

by David E. Robinson and Yue Li,  
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A project is currently being performed by GEC-Marconi Systems and the CSIRO Ultrasonics Laboratory, in conjunction with the Maritime Operations Division, DSTO in Australia to develop an ultrasonic imaging system for undersea use. The system will operate in pulse-echo mode in the nearfield, in the low Megahertz range, with a .5m aperture, and an operating range of 1 - 4 m. There is presently negligible data on the properties of the seawater environment at these frequencies. The medium aberrations possible are:

- Refraction by local macroscopic variations in sound speed causing non-alignment in time of the received echoes (after correction for focus or geometric path length).
- Scattering from microstructural variations in sound-speed or density, or particulate matter causing changes in pulse shape or phase variations in longer pulses.

Measurement hardware is being developed to carry out comprehensive data acquisition with 32 elements in a linear .5m aperture, and 128 elements in a .1m by .1m square aperture over a 2m path to a depth of 30m. In the mean-time, preliminary measurements have been made in Sydney Harbour of observed coherence of 4 MHz pulses transmitted over a path of 1 m and received by four elements across an aperture of .5 m. Two types of signal were used for the measurements. A short pulse similar to that used in medical ultrasonic imaging, was used to assess the short-term coherence. A tone burst allowed conventional cross-correlation coherence measurements to be performed to establish uniformity of propagation during the pulse. The findings from the preliminary study were:

- No evidence of forward scatter leading to multi-path propagation and pulse spreading has been observed. This is so for short pulses and long tone bursts.
- A sharp change in vertical sound-speed profile (of  $2\text{ms}^{-1}$  over 1m) did not affect the ability to image in the horizontal plane, even at the discontinuity.
- The sharp change in vertical sound-speed profile did affect the ability to image in the vertical plane, which could be modelled as a combination of vertical shift, defocussing and random blurring of the image.

The conclusion is that the predominant effect observed was refraction due to stratified temperature variations. In the conditions observed, signal incoherence would not substantially affect ultrasonic high-resolution 3-D imaging at 4 MHz.

It is planned to use the test equipment being developed to gather data at a number of locations near Darwin on the northern Australian coast in June this year. These trials will reveal in somewhat more testing conditions the nature and severity of aberrations likely to be encountered, and the extent of aberration correction algorithms required in an operational system.

The opportunity exists for international collaboration to extend the data gathering to a variety of environments. This will provide a more complete understanding of the aberrations likely to be encountered as the system is used in different geographical locations. This better understanding will have an impact on the type of aberration correction algorithms to be developed. The development of algorithms could also form part of an international collaboration effort.

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## CERAMIC PROCESSING FOR IMPROVED CONTROL OF PZT CERAMIC PROPERTIES

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One of the major difficulties which can be encountered in manufacturing PZT ceramics is the batch to batch variation in the piezoelectric and dielectric properties. Some suppliers quote a  $\pm 10\%$  production variation from the actual values stated in their data sheets. The reasons for such variations are quite numerous and may include raw material changes, mixing and grinding processes, firing conditions and poling conditions. The Sensors group within GEC-Marconi Systems, Australia, has developed a technique for tracking these variations, which we call the  $K^T-k_p$  diagram.

The  $K^T-k_p$  diagram is created by plotting the dielectric constant against the coupling coefficient for a number of titanium/zirconium ratios for the one basic PZT composition. For PZT ceramics made by the conventional mixed oxide route the phase boundary, as indicated by the maximum in the dielectric constant, occurs at a higher titanium content than the maximum in the coupling coefficient. When the results from other batches are superimposed on this trend line, it is clear that much of the batch to batch variation is caused by apparent changes in the Ti/Zr contents. This may be caused by many factors, for example, a different sintering temperature or rate may alter the stoichiometry through variations in lead losses. This causes a rhombohedral phase shift which may have a much greater effect than that of increased grain size. Other such examples and applications of the diagram will be discussed in more detail.

A further aim for GEC-Marconi has been the development of scale up principles to allow production processes to be simulated on a laboratory scale. Once such a correlation has been established, laboratory trials may be used to predict the properties achieved in a production batch.

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## NET-SHAPE PIEZOCOMPOSITE TRANSDUCERS FOR ULTRASONIC IMAGING ARRAYS\*

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Piezoelectric ceramic/polymer composites, already in use in medical ultrasound as imaging transducers operating at megahertz frequencies, are now finding application in large area arrays for undersea imaging. The medical transducer industry uses dice-and-fill methods for producing the very fine piezoelectric ceramic elements required in a typical imaging array. Although this manufacturing method has served the industry well for over ten years, future requirements for higher operating frequency and 2D layout will require extremely fine elements, improved control of interelement coupling and crosstalk, and advanced array designs that challenge the capabilities of dicing technology. Similarly, several Navy undersea acoustic imaging applications, covering a wide frequency range from sonic frequencies up to the megahertz range, will need large quantities of piezocomposite transducer materials at low cost.

Under ONR and ARPA funding, Materials Systems Inc. has developed net-shape ceramic injection molding processes for cost-effectively manufacturing complex arrays of the fine PZT ceramic elements required for advanced composite transducers. Net-shape formed piezocomposites are now becoming available in commercial quantities for the first time, allowing new transducer configurations to be developed for medical ultrasound and nondestructive testing, as well as undersea imaging, surveying, sensing, and actuation.

In this presentation, Materials Systems Inc. reviews the current capabilities of its PZT injection molding manufacturing process for fabricating various composite transducer designs relevant to high frequency medical ultrasound, undersea imaging, and nondestructive testing. Recent results from an ongoing international Technical Cooperation Program, in which MSI is collaborating with other transducer and systems design and testing groups on both sides of the Atlantic, are presented. Methods for producing complex composite element layouts using low cost tooling are discussed. Additional capabilities, anticipated to become commercially available within the next one to two years, include extremely fine PZT element dimensions ( $<25\mu\text{m}$ ), high PZT volume fraction, new polymer matrix materials, improved dimensional control, large area devices, and reduced cost.

\*Supported by the Office of Naval Research.

## EXPERIMENTAL STUDIES OF PIEZOCOMPOSITES PART II: DYNAMIC BEHAVIOR

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In Part I, we reported a comparison of predicted resonance frequencies of 2-2 piezocomposite samples with different structures and materials using three theoretical models<sup>1-3</sup> and computer modeling using finite element methods. It was shown that the thickness mode, the lateral modes and mode coupling can be strongly affected by the choice of constituent materials, and the specific structure used. This approach can be exploited to predict the required operation modes, while suppressing undesired modes.

In Part II, we report the use of interferometry to investigate the predictions of the modes. The computer controlled data collection system consists of a scanning heterodyne interferometer with the sample configured as a mirror in one of the arms. A translation stage moves the sample in a raster pattern, while a computer records the point-by point magnitude and phase of the surface displacement for later analysis. From the data, the periodic deformation of the surface can be reconstructed and displayed.

The piezocomposite samples consisted of regular periodic 2-2 and 1-3 structures, as well random-cut structures. These samples had low and high shear velocities and attenuation which were designed for the investigation of the various significant aspects of composite behavior such as displacement amplitude and phase of the ceramic and polymer.

The animations demonstrate that the vibrational character of the thickness mode differs from that of the lateral modes. These differences can be determined by comparing the magnitude and phase of the polymer and ceramic for different samples. These animations reveal all existing modes in the composite. Consequently, these animations serve to verify the basic elements of the models, while identifying their limitations as well.

Further investigation into the relationship between transducer performance and the properties of piezocomposites is under way, and will be reported as Part III.

The financial support by the ONR through the Grant No. N00014-91-C-0229 is greatly appreciated.

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# ELECTRO-MECHANICAL-THERMAL FINITE ELEMENT MODELING METHODOLOGY FOR TRANSDUCER ARRAYS

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Rigorous computer modeling of transducers is achieving its promise as an engineering design tool—augmenting the usual physical experiments and approximate (1D) analyses with robust, multidimensional numerical experiments. In this regard we review PZFlex, a workstation-based, finite element code that solves the full 2D/3D electromechanical equations in the time-domain. Methodology issues include extended 1D and 2D array models, transient thermal models, and efforts toward more comprehensive material characterization. This work is supported in part under an ONR Contract for a modeling collaboration with Prof. G. Hayward, University of Strathclyde.

Cross-talk in composite arrays is of course an important design issue. Electromechanical waves propagating laterally in the composite are dispersed by its finite thickness and periodic structure, but current theory only provides a qualitative understanding. We describe pros and cons of a “large-scale” numerical dispersion model and use it to quantify cross-talk waves, including Bragg reflection. Examples include an existing composite plate built by UDI-Wimpol, Ltd., and a preliminary 500 kHz imaging transducer design.

Thermal effects can be a limiting factor in certain situations, e.g., high-power sonar transmitters and medical transducers operating in the Doppler mode. The designer needs to quantify maximum temperatures, diffusion paths, and effects of heat sinks or increased conductivity. Under an NIH grant on ultrasound therapy the necessary transient thermal capability has been added to PZFlex. We will present some examples for composite and medical arrays.

Without good material properties, modeling loses much of its value. The burden of piezoceramic characterization is typically born by manufacturers, and their modeling success is proportional to their measurement ability. We recently collaborated with Prof. Binu Mukherjee/Mr. Stewart Sherrit of the Royal Military College of Canada and Motorola to fully characterize Motorola PZT-type materials using IEEE standard resonator samples. We will conclude by discussing early results, including measurements and resonator models.

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## FINITE ELEMENT ANALYSIS FOR A NEW COMPLEX 1 - 3 TYPE COMPOSITE MATERIAL TRANSDUCER

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In this paper, the authors applied both two - and three - dimensional finite element computer codes developed by Dr. Huang to simulate the new complex 1-3 type piezoelectric composite material transducer (a detailed description of a layered piezo composite can be found in authors' paper of 1994 IEEE Ultrasonics Symposium Proceedings). The quantum mechanics Dirac notation is used to derive a complete set of finite element constitutive equations with damping. A complex 1-3 type composite prototype transducer with a 25% volume friction is tested. Experimental measurements peak Transmitting Voltage Response (TVR), Receiving Voltage Response (RVR) and Figure of Merit (Insertion Loss) are 158.0 dB, -178.4 dB and -21.3 dB respectively. The in water mechanical quality factor, Q, is 2.5 with the -3 dB transmitting bandwidth covering from 80kHz to 125kHz. The experimental data agrees with finite element modeling results very well. The frequency domain sound pressure at observation point is calculated by the Kichhoff's integral, provided the active acoustic radiation surface particle velocity distribution defined by the finite element simulation. Assuming a delta input voltage excitation in the time domain, we may expect a flat voltage response in the frequency domain. By sweeping the frequency during the calculation of finite element constitutive equations in frequency domain and by virtue of the Kichhoff's integral, we can get acoustic pressure spectrum of the impulse response at the observation point. The time domain sound pressure waveform for any other voltage excitation can thus be easily obtained by applying inverse FFT and convolution techniques. Several time domain analysis examples for the new complex 1-3 type composite material transducer are presented.

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# THE EFFECT OF COMPOSITIONAL MODIFICATIONS ON THE PROPERTIES OF FIELD INDUCED ANTIFERROELECTRIC-TO-FERROELECTRIC CERAMICS

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Lead Lanthanum Zirconate Titanate Stannate (PLZST) based ceramics have been investigated for application in charge storage capacitors and high strain transducers/actuators. In our previous study of the compositions  $(\text{Pb}_{0.98}\text{La}_{0.02})(\text{Zr}_{0.66}\text{Ti}_{0.11-x}\text{Sn}_{0.23+x})\text{O}_3$  denoted by  $x$  on the phase diagram below, it was shown that increasing the Ti/Sn ratio resulted in a higher AFE-FE transition temperature, sharpening of the dielectric peaks ( $\epsilon(T)$ ), increase in dielectric constant (RT) and decreased switching field. It was also shown that the longitudinal strains were all about 0.2% at the phase switching field and continued to increase with increasing field level even after phase switching was completed.

In this study, the Zr/Ti ratio was extended along the phase boundary (denoted by  $\Theta$ ) in order to better understand the hysteresis behavior and switching field levels. In addition, further compositional modifications were made by substituting Ba and Sr into the A-site position of PLZTS (denoted by  $a$  and  $b$ ). Preliminary results in this study regarding the 5% Sr modifications indicated that these substitutions shift the  $\text{AFE}_{\text{Tet}}-\text{FE}_{\text{Rh}}$  boundary upward, toward the  $\text{Pb}(\text{Zr}_{0.5}\text{Ti}_{0.5})\text{O}_3$  composition. Higher switching fields with low hysteresis was observed with Sr additions. The Ba addition shifts the  $\text{AFE}_{\text{Tet}}-\text{FE}_{\text{Rh}}$  boundary downward, toward the  $\text{PbZrO}_3-\text{PbSnO}_3$  binary line. For the  $b$  composition with 5% Ba modification, FE behavior at room temperature with a transition to AFE at 40°C was noted. The longitudinal strain for this sample was 0.27% at both room temperature (FE state) and 70°C (AFE state). Field induced polarization, strain and dielectric properties were studied. The phase transition behavior will also be discussed with respect to the perovskite tolerance factor and the configuration of the  $\epsilon(T)$  curves.

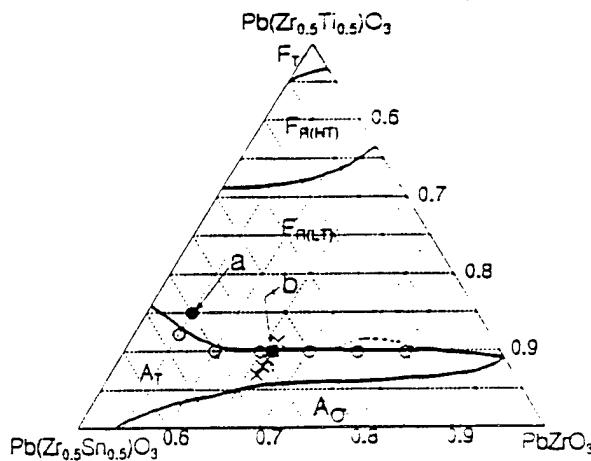


Fig. Triaxial phase diagram for the system  $\text{Pb}_{0.98-x}\text{La}_{0.02}(\text{Zr},\text{Sn},\text{Ti})\text{O}_3$

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## HYDROTHERMAL SYNTHESIS OF PEROVSKITES FOR TRANSDUCER MATERIALS

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The objective of this research was to develop a first principles approach for the hydrothermal synthesis of perovskite oxides useful for transducer materials. A thermodynamic approach has been developed capable of specifying the reaction conditions required to form a phase pure material. A series of perovskites relevant for transducer and other electronic applications were investigated and successfully modeled. These systems include titanates and zirconates of barium, strontium, calcium, and lead. In addition, the solid solution lead zirconate titanate was also investigated. Alkaline earth titanates and zirconates exhibited stability only at high pH. Excesses of alkaline earth ions will not contaminate the powder unless an excess capable of precipitating the hydroxide is present. In contrast, lead-based perovskites exhibited amphoteric stability where a low pH or excessively high pH could render the compound unstable. However, excess lead in solution can lead to a narrow set of conditions where phase-pure materials can be produced. For solid solutions such as lead zirconate titanate, the zirconium to titanium ratio was critical towards determining this pH range for perovskite phase stability. Improvements in phase yield are predicted to improve if very slight excesses of lead are employed (e.g.,  $Pb/(Ti+Zr) = 1.003$ ). Room temperature phase equilibria was found to be representative of that observed at higher temperatures on a semi-quantitative basis. All the above materials exhibited extreme sensitivity to carbonate formation and resultantly should be processed in carbon dioxide-free environments. The most challenging aspect of this research was judicious choice of thermodynamic data and development of estimation procedures in the absence of experimental data. Estimation procedures developed by others as well as ourselves will be summarized.

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## STRUCTURAL AND PROPERTY STUDIES OF HIGH ZR-CONTENT PZT

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Transmission electron microscopy (TEM) studies have been performed on  $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$  for  $0 \leq x \leq 0.2$ . These studies revealed several new important features of the phase transformational sequence. Disordered R-type and M-type oxygen rotations were found in the paraelectric state. Near the Curie temperature, an ordering of oxygen rotations was observed. For  $x < 0.05$ , bright-field imaging and convergent beam electron diffraction (CBED) patterns revealed an intermediate ferroelectric phase with rhombohedral symmetry between low-temperature antiferroelectric orthorhombic and high-temperature paraelectric cubic states. This intermediate ferroelectric state

was characterized by the presence of  $\frac{1}{2}[110]$  superlattice reflections, associated with ordered M-type oxygen rotations. Lattice imaging of the  $\frac{1}{2}[110]$  reflections revealed  $\langle 110 \rangle$ -orientated structural modulations. The lengths of the structural modulations were  $\sim 100 \text{ \AA}$ . The local symmetry due to the structural modulations was tetragonal, however averaging over the various orientations resulted in a macroscopic rhombohedral symmetry. With increasing Ti-content, the

intensity of the  $\frac{1}{2}[110]$  reflections was found to decrease, however on cooling strong  $\frac{1}{2}[111]$  reflections were found to develop. It is believed that a hierarchy of symmetries exists over a wide region of the rhombohedral ferroelectric phase field. The influence of the oxygen rotations on the macroscopic properties (including the dielectric, piezoelectric, and electrically-induced strain) will then be discussed.

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## CERAMBOWS: PRE-STRESSED COMPOSITE CERAMIC ACTUATORS

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Amplified mechanical displacement effects, similar to those observed in the recently reported Rainbow actuators, have also been found to exist in pre-stressed ceramic/metal composite structures coined as CERAMBOWs - an acronym for CERamic And Metal Biased Oxide Wafer. Mimicking the Rainbows in many ways, the intentionally created internal compressive and tensile stresses within the Cerambows are used to amplify their displacement properties via the combined effects of piezoelectric  $d_{31}$  strain and domain re-orientation. They are fabricated from ferroelectric, piezoelectric or electrostrictive materials and metal substrates of significantly different thermal expansions which are largely responsible for the creation of the stress. Typical ceramics used in Cerambows were PZT, PLZT, PBZT, PSZT and PMN; and some metal substrates tested were Al, Ag, Ni, brass, steel and Be/Cu foil in varying thicknesses ranging from 0.025mm (1 mil) to 0.25mm (10 mils). Shapes varied from round disks to square plates and rectangular bars. Formed at an elevated temperature of approximately 250°C, the stresses on cooling to room temperature were generally sufficient to produce displacements as large as 0.125mm (5 mils) when activated unipolar and 0.25mm (10 mils) when operated bipolar at 450 volts in a dome mode. Comparing equal structures of a Cerambow with a Rainbow, the Cerambow was found to achieve approximately 70% of the displacement that would normally be obtained with a Rainbow. Although this difference in displacement is sufficient to prefer a Rainbow for many applications, there are some advantages for the Cerambow. Among these are (1) the processing temperatures are lower, (2) high lead-containing ceramics are not required and (3) in some instances the metal substrate is more convenient to interface with other elements of a device. However, the disadvantages include (1) lower displacement in the dome mode of operation, (2) the higher displacement saddle mode has not yet been demonstrated with a Cerambow and (3) the ceramic/metal bond interface is a possible failure area when operated for extended periods of time. The applications for Cerambows are considered to be similar to Rainbows, i.e., actuators, pumps, deflectors, vibrators, speakers, hydrophones, hydroprojectors, switches, etc.

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***Poster Summaries***

## MULTI-BEAM, ACOUSTIC LENS IMAGING SONAR

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Since acoustic waves pass relatively unscathed through turbid water, high resolution sonars can image objects in conditions that obscure underwater video, photographic, and laser systems. This presentation describes and evaluates an experimental 3 MHz sonar that forms 96 beams, each 0.25° in azimuth by 7° in elevation. Two thin, rectangular lenses determine the azimuthal beamwidth. The transducer element curvature and diffraction slit determine the elevation beamwidth. No electronic beamforming is required. The array mechanically shifts 3.5 mm left and right to obtain two sets of interleaved beams which together generate the 96 beam b-scan image. The sonar has 0.3 cm down-range resolution and 1 cm cross-range resolution when imaging objects at a 3 m range. The 48 transducer elements are ceramic stacks each 2 mm wide and 30 mm long and are made of 1-3, 50% volume fill composite comprised of PZT and epoxy filler. The curved backing material, front matching layers, and low impedance material between the elements improve performance. The p-p insertion loss of the elements averages -53 dB. The elements were tuned to 3 MHz and have an average of a 78% bandwidth (to the -6 dB points). The electrical crosstalk in the retina between adjacent elements was less than the acoustic crosstalk which varied from -45 dB to -58 dB. The electrical crosstalk in the sonar system (retina not attached) measured -76 dB for channels that did not share a common transmitter and -60 dB otherwise. High resolution images were formed on a Silicon Graphics workstation at frame rates of 1.2/s or higher depending on image size. These images showed bolt patterns and other small features of targets of interest that would allow target identification even in turbid water.

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## PIEZOELECTRIC COMPOSITE TRANSDUCER ARRAYS FOR UNDERWATER IMAGING SYSTEMS

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A review of undersea ultrasonic imaging applications has been conducted to determine the range of potential uses for electronically scanned imaging systems as part of The Technical Cooperation Program (TTCP) sponsored by the US Office of Naval Research and the UK Defense Research Agency. The review was conducted to determine the basic imaging system performance parameters required to meet the requirements of detection, classification, and identification of underwater objects. From these basic parameters, specifications for transducer arrays to support new imaging systems are being developed. These specifications in turn will determine the requirements for fine-scale piezoelectric composite materials to support these arrays.

A Mine Counter-Measures (MCM) application was chosen as the focal point for the development effort. As in most sonar applications, separate projector and receiver arrays are employed in order to separately optimize the transmit and receive signal paths. The transducers are designed with diverging fan beams slanted along the elevational axis to sort out returning echoes in range. This paper will present the design of a receiver array in both in its beamforming characteristics and preliminary composite microstructure. A curved array design was chosen over a phased array design to minimize beamformer cost and to achieve acceptable angular beamwidth in a monolithic composite structure.

The curved receiver array design parameters were governed by the necessity of confining the size of the array to 235 mm in length. This allows the array to fit inside a present MCM vehicle head for test purposes. This paper will show the development of the basic array specifications based on the requirements of 1.5° far-field beam width, a 90° field of view with full aperture active on all scan lines, and a maximum assumed acceptance angle for the array of +/-30°.

The composite microstructure of the array will be the subject of research over the next year. Based on preliminary studies conducted by collaborators at Strathclyde University, Weidlinger Associates, The Royal Military College (Canada), UDI-Wimpol, and Materials Systems, the initial design of the composite array will be presented. This design will be refined and prototype arrays will be constructed over the next twelve months.

## MICROMACHINED PZT HIGH FREQUENCY SONAR TRANSDUCERS

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Millimeter sized ferroelectric unimorph sonar transducers have been built using micromachined sol-gel PZT on silicon wafers. Arrays of transducers have been tested in water in the frequency range 0.5 to 4 MHz. Potential applications for these transducers are high frequency imaging sonars, medical ultrasound, and flaw detection (NDT).

Transducers were fabricated on 3" silicon wafers using standard micromachining techniques. Individual transducer diaphragms varied from 0.2 to 2 mm in size. Boron diffusion was used to form an etch-stop membrane of controlled 5 or 10  $\mu\text{m}$  thickness. These membranes were coated with silicon dioxide insulator and Ti/Pt metal bottom electrodes. The wafers were then coated with Sol-Gel PZT at Penn State MRL. The top Pt electrodes were then deposited and patterned using a lift-off procedure, and the PZT layer was patterned using wet chemical etching.

The Sol-Gel technique has previously been widely used to deposit thin-films of PZT (< 1  $\mu\text{m}$  thick) on silicon wafers for ferroelectric memories (FRAM's). This work reports for the first time high quality crack-free PZT films up to 8 microns in thickness, which leads directly to higher sensitivity and figure of merit for most transducers. The longitudinal piezoelectric coefficient  $d_{33}$  is 140-180 pC/N, a magnitude close to that of a bulk ceramic. Remanent polarization of 34  $\mu\text{C}/\text{cm}^2$ , a coercive field of 42 kV/cm, and dielectric constant  $\epsilon_r$  equal to 1000 - 1300 were measured on these films.

Results of in-water testing are presented including sensitivity, frequency response, beam patterns and output capacitance.

The authors would like to thank the Naval Explosive Ordnance Disposal Technology Center and the C. S. Draper Laboratory for supporting this work.

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## EXPERIMENT STUDIES OF PIEZOCOMPOSITES PART I: COMPARISON WITH THEORETICAL MODELS

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The most common class of piezocomposite, in which the structure is strictly periodic, has undesirable resonances associated with the lateral composite dimensions which limit the upper frequency range of operation. Seeking an approach to break this frequency barrier will benefit the Navy, and also will be very meaningful to improve the transducer performance for civilian application.

By changing the properties of the constituent phases and the composite dimensions, the properties of piezocomposites can be varied over a wide range. Both analytical and finite element modeling for piezocomposites have been carried out and provide useful guidance in transducer designing.

Experimental studies of piezocomposites have been conducted by designing and developing different types of 2-2 and 1-3 composite samples and analyzing their properties. Polymer matrix materials with low and high shear velocity and attenuation have been used to investigate the effects on the lateral resonances. Periodic and random structure have been employed to study the effects on the lateral mode suppressing.

The results of our experimental research will be presented in three portions. With Part I, the measured frequencies of resonance in 2-2 composite samples with different structures and materials are compared with previous stopband theory.<sup>1</sup> These measured resonances, along with the thickness coupling factor  $k_t$ , are also compared with dynamic model.<sup>2</sup> The predictions calculated by FEM are presented and compared to the measured data from the samples in which the matrix materials have high shear attenuation. As for the composites with random-cut structure, the comparison with predicted resonances by T-matrix method<sup>3</sup> is performed. It is shown that the thickness mode, lateral modes and their mode coupling can be deeply affected by the composite materials and structure. This approach can be exploited to predict the required modes of operation, while the undesired modes are suppressed.

The dynamic behavior of piezocomposites will be presented separately as Part II in this meeting.

The financial support by the ONR through the Grant No. N00014-91-C-0229 is greatly appreciated.

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3. W. W. Cao, W. K. Qi, Q. M. Zhang and L. E. Cross, Computational Studies on Piezocomposite Transducers. 1994 ONR Transducer Materials and Transducers Workshop. April, 1994

**ARBITRARY LINEAR, PLANAR AND SPATIAL ACOUSTIC  
ARRAY DESIGN AND SYNTHESIS:  
COMPUTER AIDED DIRECTIVITY, INSTANT VISUALIZATION  
AND EXPERIMENTAL VERIFICATION**

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In this paper, the authors applied the fast algorithm [Huang and Boucher, J. Acoust. Soc. Am. Vol.95, 1994] to design and to synthesize arbitrary linear, planar, arc, cylindrical, spherical and any other spatial acoustic arrays. The array design and synthesis computer code is written in FORTRAN language by Dr. Huang. The acoustic radiation field of the elements in the array is calculated by the Kichhoff's integral first. By extensively applying the concept of superposition, we are not only able to use the element local radiation field as an input to form the first order subarray directivity, but also able to model higher order complex system array directivity quickly. The elements involved in the array can have arbitrary weighting functions with either regular shape (disk with or without central hole, rectangular, elliptic, concentric strip ring etc.) or irregular shape (special aperture with boundaries defined mathematically or numerically: Gaussian, cosine etc.). The particle velocity distribution on the surface of the array element can be uniform or non-uniform and is calculated using a two - or three - dimensional finite element analysis. The methodology for this finite element analysis in the frequency or time domains can be found from authors' other papers in this 1995 ONR workshop and in 1994 IEEE Ultrasonics Symposium Proceedings. These elements in the array can be located anywhere in the system and be oriented in any direction in the space. Further, the acoustic array can also be in steering mode during the simulation. The baffle effect is modeled numerically by introducing a baffle function. The designed array beam pattern can be visualized and printed instantly in regular polar, conic polar, contour, three-dimensional cylindrical and spherical coordinates formats. Various array design and synthesis numerical and experimental examples will be discussed.

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DESIGN AND FABRICATION OF A FLEXI-DISTORTIONAL  
PIEZOELECTRIC SENSOR

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A composite, flexi-distortional, pressure sensor with potential for improved sensitivity and  $d_{35}g_h$  performance has been proposed by Carlson<sup>1</sup>. The work of this thesis was to design and fabricate an ambient (one atmosphere) pressure, flexi-distortional sensor, and to identify critical design and testing parameters. This included forming and poling of the PZT elements and assembly of a sensor, for which the effective  $d_{35}$  piezoelectric charge coefficient could be determined. In order to activate and amplify the  $d_{35}$  coefficient, the ability to pivot and mechanical continuity of the cap-electroceramic connection were important. Weak points in the design of these "pre-prototype" sensors were the cemented joints and low precision caps. Although the sensors were crude some success was achieved in activating and amplifying the superior  $d_{35}$  mode.

<sup>1</sup> W. B. Carlson, W. A. Schulze, R. E. Newnham, L. E. Cross, "Flexi-Distortional Piezoelectric Composites with Improved  $d_{35}g_h$  Response," New York State College of Ceramics, Alfred University, Alfred, New York, (1993-preprint).

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## ANTIFERROELECTRIC PHASE-SWITCHING THIN FILMS

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Thin and thick film antiferroelectrics are potentially attractive as hysteresis-free actuators in applications such as latching relays or air acoustics transducers. In this work,  $\text{PbZrO}_3$  - based antiferroelectric thin films have been prepared both by reactive co-sputtering and by laser ablation to study their dielectric and electromechanical properties.

Sputter-deposited films were deposited from metal Pb and Zr targets in an  $\text{Ar}/\text{O}_2$  atmosphere at a pressure of 10 - 20 mtorr. Films were deposited onto unheated rotating Pt-coated Si substrates and crystallized ex-situ using a rapid thermal annealer. Stoichiometry was controlled by adjusting the power to the individual magnetrons for the Pb and Zr sources. Good crystallinity of the perovskite phase was achieved for films deposited at a Pb/Zr power level near 20/250 and annealed at 750°C for 10 seconds. Double hysteresis loops with well-defined coercive fields corresponding to the phase switch from the antiferroelectric to the ferroelectric phase were observed for the  $\text{PbZrO}_3$  films at room temperature. The saturation polarization was  $\sim 38\mu\text{C}/\text{cm}^2$ . The dielectric constant and dielectric loss were also comparable to values expected from bulk ceramics.

Results on laser-ablated films deposited both at room temperature and at elevated temperatures will also be presented.

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## **Kinetics of Hydrothermal Lead Titanate Powder Formation**

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The kinetics of hydrothermally synthesized lead titanate was investigated in order to optimize the reaction conditions. The effects of temperature, pH, concentration, ionic strength, surface area of titania, stir rate, volume fill, and order of addition were systematically studied. A 10-20 times increase in reaction yield resulted from an increase in feed stock surface area of titania from 10-500 m<sup>2</sup>/g. A 4 fold increase was found by changing the temperature from 140-160 °C. A 1.3-2.3 times increase in the reaction yield resulted from altering the pH from 9 to 10. It was also found that the early stages of the reaction were accelerated by stirring. It can be concluded that surface area of titania source plays the most significant role in controlling the reaction rate.

## PLASMA-SPRAYED LEAD ZIRCONATE TITANATE - GLASS COMPOSITES

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A plasma-spray process has been used to produce piezoelectric lead zirconate titanate (PZT) - glass composite thick films. The starting powders consisted of mixtures of Navy Type V PZT and lead aluminosilicate glass. Three compositions were tried and these had PZT contents of 92, 85 and 77 volume percents. The powders were wet milled in methanol for 1 hour, calcined at 500 to 700 C and then remilled before being used for plasma spraying; the particle sizes varied between 1 and 3  $\mu\text{m}$ . For comparison, a few discs were pressed at 10 000 kPa with a poly vinyl alcohol binder and fired at 750 C.

X-ray diffractograms showed that the plasma-sprayed films had the same crystal structure as the PZT (Navy Type V) and lead-based glass starting powder mixture. The films showed good adhesion to stainless steel and silver coated glass slides and poor adhesion to aluminium substrates. The film thickness ranged from 0.08 to 0.25 mm and the film densities were between 2500 and 3700 kg/m<sup>3</sup>. The low film densities suggest that pores took up 30 to 40% of the volume. The dielectric constant of the films varied between 58 and 20 with dissipations between 0.019 and 0.032. The films were poled, and their piezoelectric charge coefficient,  $d_{33}$ , was found to be 1.1 pC/N.

Our work has shown that the plasma-spray process can produce piezoelectric films. However processing methods will have to be improved to increase the film densities and its microstructure in order to obtain a significant piezoelectric effect.

We thankfully acknowledge financial support from the Defence Research Establishment Atlantic, Department of National Defence, Canada.

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**The Effects of Chemical Processing Variables on Powder  
Characteristics of Hydrothermal PZT Powder**

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The effect of stirring during the hydrothermal synthesis of PZT was investigated.  $\text{Pb}(\text{C}_2\text{H}_3\text{O}_2)_2 \cdot 3\text{H}_2\text{O}$ , KOH, and hydrous  $(\text{Zr},\text{Ti})\text{O}_2$  were used as feedstock materials in the reaction at 160°C for 72 hrs. The resultant PZT powders were analyzed for Zr, Ti and Pb content. A t-test statistical analysis at 95%, 99% and 99.5% levels of confidence showed there was no difference in the Zr and Ti content between  $(\text{Zr},\text{Ti})\text{O}_2$  feedstock and the PZT powder due to stirring. However, results for the Pb content of the powders showed a difference between stirred and non-stirred reactions.

## ORIGIN OF LARGE PORES IN TAPE CAST PZT

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Single layers of tape cast PZT were examined to determine how various processing parameters affect the number density of 'very large' (>20 micron) pores. According to one manufacturer, pores of this magnitude can be detected in all commercial PZT materials, but in densities as low as a few per square centimeter. The presence of these pores can cause problems in multilayer capacitors and actuators where high electric fields cause dielectric breakdown. Statistical experimental design was effectively used to investigate the effect of several variables. Factorial and partial factorial designs allowed for the study of main effects as well as some factor interactions. Parameters that were examined include slurry milling time, filtering, deairing, composition, tape thickness, and sintering schedule. All tape casting suspensions were prepared by a two stage ball milling process of a nonaqueous slurry. Tapes were sintered in air at 1250 °C. Pores were counted by visual inspection of both surfaces of sintered tape samples under a transmitted light microscope. Statistical analysis of pore counts and sintered densities, summarized using ANOVA tables, showed which parameters had the largest effects. For instance, it was determined that deairing time and fish oil content in the composition had great impact on the formation of large pores on the surface of the tape cast PZT. An interaction between the first milling stage time and the sintering time was shown to affect the sintered density. Further experimental designs were used to optimize these parameters.

**TITLE:** PROCESS AND PROPERTIES OF LEAD TITANATE FOR 0-3 COMPOSITES

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Past zero three composites that contained lead titanate (PT) as the active phase tend to break down at low poling fields. As a result they exhibited a low piezoelectric sensitivity. The present work is addressed to the preparation and characterization of large PT particulates in which high resistivities allow the application of strong poling fields. Heat treatment conditions that provide crystallite sizes to  $100 \mu\text{m}$  and dopant levels that appear to enhance the resistivity have been observed. Conventional powder characterization methods as well as specially designed fixtures are being evaluated to determine electrical and piezoelectric properties of PT particulates. Test results indicate that there is a relationship of resistivity to post firing powder treatment, including application of humidity and voltage and of packing conditions. The properties of actual 0-3 composites will be used for validation of the results.

# INTERACTION OF AG/PD METALLIZATION WITH LEAD OXIDE, AND LEAD-BASED ELECTROCERAMICS

W. HUEBNER

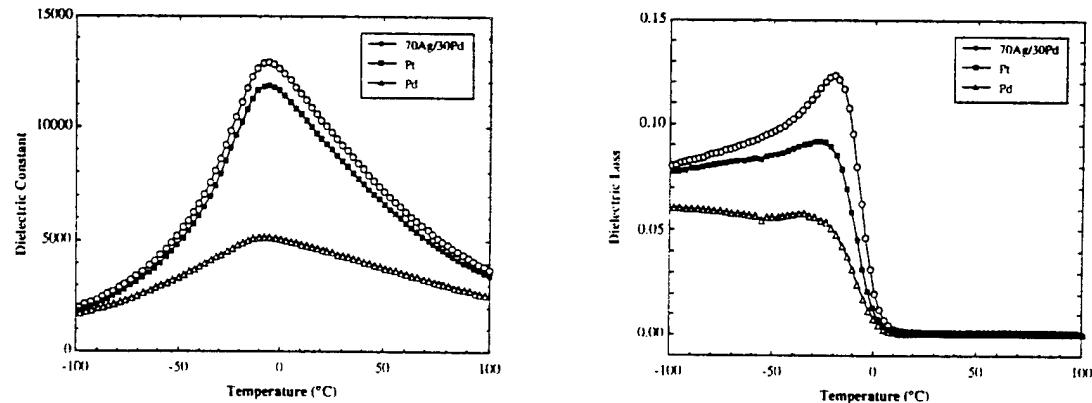
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An ongoing challenge for future developments in high performance ceramic multilayer capacitors, actuators, and integrated ceramics is to reduce the internal electrode cost and thickness without sacrificing yield or reliability. Key to these developments is a thorough understanding of the interactions which occur between flux-sintered dielectrics and low cost, Ag/Pd electrodes. In this study the phase relations between Pd or 70Ag/30Pd electrode systems with PbO, and commercially-important Pb-based oxides were determined to establish the conditions under which detrimental interfacial chemical reactions may occur. Results show the equilibrium phases which form strongly depend upon the Ag/Pd ratio and temperature. These phases include  $PdPbO_2$ ,  $Pd(Pb)$  and  $PbPd_3$ . The  $PdPbO_2$  phase decomposes when  $PdO$  destabilizes, resulting in a series of reactions which result in oxygen evolution, and partial melting of components.

For the reaction of Pd with Pb-compounds, a  $Pd(Pb)$  alloy formed which in all instances exhibited the maximum solubility of the Pb, i.e. 14 at%. These reactions have an adverse effect on the local stoichiometry and composition of the dielectric, causing depletion of Pb. Studies on  $Pb(Mg_{1/3}Nb_{2/3})O_3$  showed the use of Pd electrodes decreased the dielectric constant substantially (see figure below), which was due to the formation of a low permittivity phase in series connectivity with the unaltered dielectric.



Dielectric constant and loss of  $PbMg_{1/3}Nb_{2/3}O_3$  cofired with Pt, Pd, and 70Ag/30Pd electrodes.

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# ELECTRO-MECHANICAL-THERMAL FINITE ELEMENT MODELING METHODOLOGY FOR TRANSDUCER ARRAYS

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Rigorous computer modeling of transducers is achieving its promise as an engineering design tool—augmenting the usual physical experiments and approximate (1D) analyses with robust, multidimensional numerical experiments. In this regard we review PZFlex, a workstation-based, finite element code that solves the full 2D/3D electromechanical equations in the time-domain. Methodology issues include extended 1D and 2D array models, transient thermal models, and efforts toward more comprehensive material characterization. This work is supported in part under an ONR Contract for a modeling collaboration with Prof. G. Hayward, University of Strathclyde.

Cross-talk in composite arrays is of course an important design issue. Electromechanical waves propagating laterally in the composite are dispersed by its finite thickness and periodic structure, but current theory only provides a qualitative understanding. We describe pros and cons of a "large-scale" numerical dispersion model and use it to quantify cross-talk waves, including Bragg reflection. Examples include an existing composite plate built by UDI-Wimpol, Ltd., and a preliminary 500 kHz imaging transducer design.

Thermal effects can be a limiting factor in certain situations, e.g., high-power sonar transmitters and medical transducers operating in the Doppler mode. The designer needs to quantify maximum temperatures, diffusion paths, and effects of heat sinks or increased conductivity. Under an NIH grant on ultrasound therapy the necessary transient thermal capability has been added to PZFlex. We will present some examples for composite and medical arrays.

Without good material properties, modeling loses much of its value. The burden of piezoceramic characterization is typically born by manufacturers, and their modeling success is proportional to their measurement ability. We recently collaborated with Prof. Binu Mukherjee/Mr. Stewart Sherrit of the Royal Military College of Canada and Motorola to fully characterize Motorola PZT-type materials using IEEE standard resonator samples. We will conclude by discussing early results, including measurements and resonator models.

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**6 April 1995**

***ABSTRACTS***

# GROWTH AND CHARACTERIZATION OF RELAXOR FERROELECTRIC SINGLE CRYSTALS<sup>†</sup>

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Relaxor ferroelectric single crystals of  $(1-x)(Pb(Zn_{1/3}Nb_{2/3})O_3 \cdot x (PbTiO_3))$ ,  $Pb(Sc_{1/2}Nb_{1/2})O_3$ , and  $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3 \cdot x PbTiO_3$  were grown using the flux solution method. Various techniques were employed to increase size and yield of optically transparent crystals, including variation of the flux composition, cooling rate, growth front traversal speed and the size of the temperature gradient at the growth front. The effect of these variables on crystal quality and dielectric properties will be discussed. In addition, piezoelectric properties of selected crystals will be presented in relation to their respective morphotropic phase boundary(s).

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## ADVANCES IN THE MODELING AND MANUFACTURING OF TRANSVERSELY ALIGNED PIEZOELECTRIC FIBER COMPOSITES

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Piezoelectric fiber composites have been introduced as a means for orthotropic actuation that address the strength, reliability, and conformability issues associated with the use of monolithic piezoceramics. Fine piezoceramic fibers aligned in-plane provide the predominant composite stiffness, strength, and actuation, while a passive epoxy matrix serves to protect the ceramic, and provide load sharing among the fibers. Previous work had described several micro electromechanical models that predict the effective composite properties, and given comparisons to manufactured test articles. More recently, work has focused on improving performance of the fiber composites through novel electrode geometry, and incorporating these active composites into host graphite/epoxy plies for structural actuation applications.

Improving the actuation capability of piezoelectric fiber composites has been accomplished using an interdigitated electrode pattern. This arrangement introduces the electric field into the plane of the structure, so that the primary piezoelectric effect is utilized. Furthermore, the geometry increases the effective ceramic volume fraction, thus further increasing the free-strain actuation. The piezoelectric free-strain constant has increased on the order of five times, as compared to the conventional electrodes. A peak strain of 1000 ppm (1 Hz, 60 °C) is possible with the new composites, a large improvement from the previous specimens. These experimental measurements of composite properties correlate well with models.

To illustrate the capability of piezoelectric fiber composites in structural actuation applications, an adaptive composite plate [90/45<sub>A</sub>/0/-45<sub>A</sub>/90] with embedded conventional fiber composite actuators has been manufactured and tested. The actuators were produced in a conformable prepreg form, embedded and co-cured along with host graphite/epoxy plies. The twist and bend induced in a 15 cm by 6 cm plate was measured and compared with predictions from a micro electromechanical model of the active plies and a Rayleigh Ritz plate model. Classical Laminated Plate Theory was used to determine stiffness properties of the laminate and the actuator forces and moments. Actuating a single active ply induced 1.2 MPa of shear stress, causing 3 deg/m of induced twist and 2.5 deg/m of induced bending in the length direction. The models were able to predict the general response; however, unmodeled frequency-dependent effects and material property uncertainties reduced the accuracy. Better material characterization and improvements in the manufacturing process will further increase performance levels in future test articles.

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## Electric Field Forces as a Means to Develop Processing of New Materials

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Electric fields, when applied to suspensions of particulate materials, permit changes in the spatial particle distribution. This redistribution of particles can be used as a means to consolidate and assemble particles into thick film structures and particle-polymer composite connectivity. The phenomena being considered to aid the processing are known as electrophoresis and dielectrophoresis.

First, dielectrophoresis has been considered as a means to form quasi 1-3 composites. Optimum assembly conditions are found for a variety of systems and discussed in terms of dielectro-viscous properties of the particle-polymer components. Dielectric properties are discussed in terms of mixing law modifications and observed microstructures.

Second, electrophoretic deposition has been investigated as a means to consolidate submicron particles for thin/thick films (1 to 10  $\mu\text{m}$ ) on  $\text{Al}_2\text{O}_3$  substrates. The densification of the electrophoretic deposited films is discussed and correlated to the dielectric and microstructural properties. The results on  $\text{BaTiO}_3$  could be expanded into other materials and the possible impact to the piezoelectric industry will be discussed.

## MOONIES, CYMBALS, AND BBs

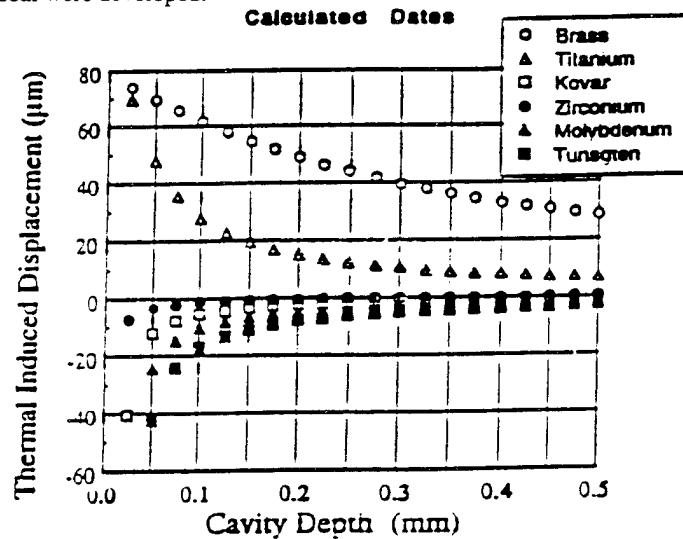
R. E. NEWNHAM\*, A. DOGAN, J. T. FIELDING, J. F. FERNANDEZ, D. SMITH  
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The unique design of the flexextensional ceramic-metal composite transducer, or "moonie" exhibits large displacements and generative forces. A new design, the "cymbal", improves properties by combining flexextensional and rotational motion of the end cap. From the previous analysis of the moonie a new, simplified model has been developed for cymbals. The effects of different metal end caps and different piezoelectric ceramics as driver elements in the system have been investigated. The results of the present work are highlighted, including displacement of 40  $\mu\text{m}$  for 2 mm actuators with loads as high as 80 Newtons for such a system. The results are correlated with the properties of the metal and the ceramic on the basis of the model.

Temperature induced displacements caused by differential thermal expansion can be a problem with these composite actuators. As shown in the figure below, the displacement of the cymbals can be controlled over a range of temperatures by selecting cap metals with the appropriate thermal expansion coefficient.

The effect of utilizing a multilayer stack in reducing drive voltage for large displacements has been demonstrated. The effect of the different end cap metals and the frequency response and displacement of the cymbal has been correlated with the stiffness of the metal. A data base catalog with bonding and asymmetry defects was established based on the analysis of the resonance spectra. Hydrostatic properties are excellent for both the cymbal and the moonie systems. Cymbals have a higher pressure sensitivity than the moonie, reaching figures of merit of  $10^6 \times 10^{-15} \text{ m}^2/\text{N}$ .

Processing and characterization studies were also conducted to optimize PZT hollow spheres for hydrophone applications. For radially poled transducers, the principal breathing mode resonance was  $\sim 700$  kHz. Hydrostatic  $d_h$  coefficients  $\sim 1,000$  pC/N were measured. Improved techniques for applying the internal electrode and epoxy seal were developed.



Calculated thermal induced displacement of the cymbal due to thermal expansion mismatch between the ceramic and the metal end cap for a 50°C temperature range.

**6 April 1995**

***Poster Summaries***

## ACTIVE CONTROL OF SOUND WITH FOAM-PVDF COMPOSITE TRANSDUCERS

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In this paper we will discuss the development and application of foam-PVDF composite transducers for noise reduction. The transducer consists of curved layers of PVDF piezoelectric film embedded in partially reticulated polyimide foam and is designed to provide both active and passive damping of acoustic waves in air. An analytical model of the transducer has been developed and will be used to illustrate the use of the material in active control of reflection of sound waves. Different configurations of the transducer have been studied and tested for the best arrangement. In particular the geometric shape and the spatial polarity distribution of the PVDF layer has been shown to be important. The results of experiments on active control of both sound radiation and reflection using the foam-PVDF in conjunction with a feedforward LMS controller will also be presented. [Work supported by NASA LaRC and the ONR.]

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## COMPOSITE SMART MATERIALS FOR DEFENSE AND DUAL-USE APPLICATIONS

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The Composite Smart Materials program is one of a number of smart materials initiatives funded by ARPA. ONR is acting as ARPA's agent for the program which is focused on developing a fully integrated smart material containing actuators, sensors, conditioning and processing electronics, and power supplies for use in underwater applications. The team members include AVX Corporation, Virginia Polytechnic Institute (VPI), Virginia Power Technologies (VPT) and the Naval Research Laboratory (NRL). Martin Marietta Laboratories • Baltimore (MML•B) is the prime contractor for the program.

The approach being taken is to design and develop an active layer using actuators and sensors fabricated on high-volume production lines used for manufacture of surface-mount capacitors, and to lay these down on printed circuits laid out on Kapton or similar materials using "pick-and-place" machines in the same manner as would be found in high-volume microelectronics production. This task is being done primarily by AVX. On the back side of the active layer, the electronics associated with signal conditioning, beamforming, and acoustic analysis would be placed, using techniques developed for high-density interconnect packaging first developed at G.E. Corporate Research and Development Laboratories under ARPA (previously DARPA) funding, and currently under further development at Martin Marietta Laboratories • Baltimore.

VPT is developing miniaturized power supplies that will be incorporated on the backplane along with signal conditioning and processing electronics. Power demands for the system may run as high as 200 W/in<sup>3</sup>. The Naval Research Laboratory and Martin Marietta Laboratories • Baltimore are developing analytical and modeling tools to help design of the composite and prediction of its performance, while VPI is developing and applying advanced fiber-optic sensor technology for health monitoring of the composite and environmental sensing. The composite will be assembled and tested at Martin Marietta Laboratories • Baltimore

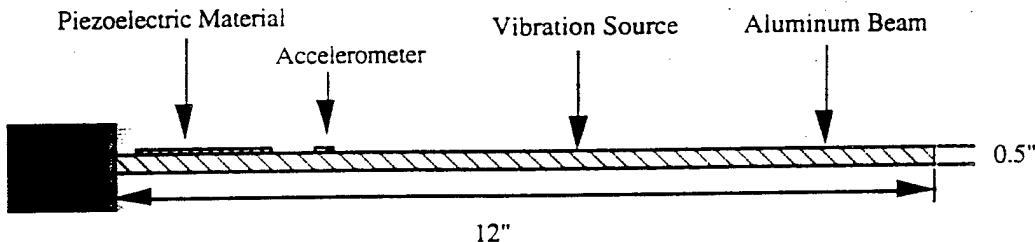
Operational requirements, design approach and preliminary test results for an early prototype of the active layer will be presented.

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## EVALUATION OF PIEZOELECTRIC COMPOSITE AS SENSOR/ACTUATOR COMBINATION IN VIBRATION CONTROL

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Piezoelectric composite materials were prepared using lead zirconate titanate (PZT) ceramic powder and epoxy. The obtained composite sample was surface-bonded on aluminum test specimens. The utilization of this PZT/epoxy composite as sensor and actuator for vibration suppression of flexible structure was evaluated and the results are compared to that of PZT.



## A CONCURRENTLY ENGINEERED ADAPTIVE COMPOSITE PANEL

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In the past decade, a significant amount of research has been devoted to the development of piezoelectric actuators capable of actively controlling structural borne vibrations that produce radiated sound power. Generally, these piezoelectric actuators are attached to the surface of or embedded in a host material structure. In many of these applications; however, the piezoelectric configurations generate insufficient levels of out-of-plane displacements of the control surfaces of the structure to control its radiated sound in the low frequency ranges. In this paper, the design, fabrication, and demonstration testing of a novel, adaptive, quiet composite, sandwich panel which addresses this problem is presented. The composite panel consists of inner and outer composite laminate skins separated by an array of cascaded flexextensional actuators embedded in a structural grade core material. Each flexextensional actuator in the array is driven in its 33 mode by a multi-layered, co-fired piezoceramic stack. The resulting surface vibrations produced by the adaptive panel are capable of reducing radiated sound power by 10 to 20 dB.

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## CERAMIC-METAL COMPOSITE TRANSDUCERS FOR HYDROPHONE APPLICATIONS

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In addition to their actuating capabilities, the "moonie" and "cymbals" ceramic-metal composite transducers can also be used for hydrophone applications. These composites are characterized by a very large figure of merit ( $d_h \cdot g_h$  or  $d_h \cdot g_h / \tan\delta$ ), high sensitivity, low dielectric loss, and great mechanical strength (i.e. high pressure tolerance). To enhance the  $d_h$ , these transducers convert a portion of the z-direction stress into large radial and tangential stresses of opposite sign, thereby causing the  $d_{33}$  and  $d_{31}$  contributions to  $d_h$  to add rather than subtract ( $d_h = d_{33} + 2d_{31}$ ). The composite design also eliminates bending stresses which might otherwise fracture the ceramic. Under hydrostatic pressures up to 1000 psi ( $\approx 7$  MPa), the "moonie" and "cymbals" designs maintain  $d_h$  values up to 1700 and 5000 pC/N and  $g_h$  values between 65 and 200 ( $\times 10^{-3}$  V·m/N), respectively. Thus figures of merit between  $10^5$  and  $10^6$  ( $\times 10^{-15}$  m<sup>2</sup>/N) are achievable.

## THE EFFECT OF DESIGN ON THE CHARACTERISTICS OF THE "MOONIE AND CYMBAL" ACTUATORS

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### ABSTRACT

Moonie type actuators fill the gap between multilayer and bimorph actuators. The effect of the geometry on the actuator characteristics of the moonie were studied. The cavity beneath the endcap plays a crucial role on the moonie actuator performance. The effect of the cavity size on the moonie characteristics were investigated. Displacement increases rapidly with increasing cavity diameter and increases linearly with increasing cavity depth. However, highly position dependent displacement characteristic and low generative force of the moonies are significant disadvantages for certain applications. The moonies were modified systematically using Finite Element Analysis and compared with experimental results. A new actuator design (the "Cymbal") was developed which gives larger displacement and generative forces along with cost effective manufacturing. The cavity of the cymbal endcap has a truncated conical shape. A die punch was designed to fabricate identical endcaps with minimal cost. Cymbal actuators show higher displacement values of about 40  $\mu\text{m}$ , with less position-dependent behavior. They also have higher generative (blocking) forces (15 N) due to the enlarged active surface, and lower metal content.

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## THE EFFECT OF MATERIALS ON THE PERFORMANCE OF THE "CYMBAL" ACTUATOR

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### ABSTRACT

Metal-ceramic composites with 2 (0)-2 2 connectivity have demonstrated very good actuator properties with moderate displacements and generative forces. The principal advantages of the new actuator design are its small size and simplified construction. At the moment, several parameters related to the performance of the actuator are not thoroughly understood. In this study the effect of the stiffness of the metal endcaps and the piezoelectric coefficient of the PZT ceramics on the performance of the cymbal actuators were investigated. The properties of the cymbal actuator that were studied are displacement, hysteresis, blocking force, and resonance spectrum. In previous studies it has been reported that the thermal expansion coefficient difference between the PZT ceramic and brass the endcaps causes a large thermally induced displacement. By selecting a metal with appropriate thermal expansion coefficient it is possible to overcome this problem. Calculated results showed that the cymbal actuators with Kovar (iron-cobalt alloy) endcaps have minimal (almost negligible) thermally induced displacement. The driving voltage of the actuator was also reduced by using a multilayer ceramic stack.

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## PIEZOELECTRIC COMPOSITE MATERIALS FOR SENSOR/ACTUATOR COMBINATIONS

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A new approach towards preparation of piezoelectric PZT/epoxy composite materials has been developed. The piezoelectric ceramic phase loading in the composite was increased up to 85 volume percent. As a result of the high PZT volume fraction in the composite, both dielectric and piezoelectric properties are improved. The conformable characteristic and the sensor/actuator abilities of this material make it useful in application of sensor/actuator combinations. A modeling of this composite was proposed to explain the structure-property relations of the composite.

## VIBRATION ISOLATION USING PIEZOELECTRIC TRANSDUCERS

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Using three Rainbow transducers and a Kistler 5-g accelerometer, we have built a system which greatly reduces levels of vibration on a load which is to be isolated from vibration of a platform. The platform itself is driven by three other Rainbow transducers. Presently the accelerometer monitors the amplitude and phase of the load vibration, which is minimized by manually adjusting the amplitude and phase of the vibration-isolating transducers. Next we will use a second accelerometer on the platform to provide a nonfeedback damping signal to eliminate the greater part of the vibration, and employ the accelerometer on the load to eliminate most of the remaining vibration by means of a feedback system such as that described below.

We are analyzing such a feedback system in which signals derived from an accelerometer are converted to acceleration, velocity, and position (PID) signals which are fed back to piezoelectric transducers whose function it is to minimize load displacement and acceleration. One feature of this feedback is the time delay between obtaining the information and using it to modify the driving voltage of the transducers at discrete time intervals. Another feature is a "forgetting function" which gradually lets the load forget its initial position, thus enabling it to adjust gradually to a new platform position. The discrete nature of the feedback process allows the possibility of chaotic behavior for certain feedback parameter combinations, but so far we have not observed chaotic behavior.

We developed a discrete sampling model and put the resulting equations in matrix form. These matrix difference equations were put on MATLAB to obtain the long-time response of the system. We also derived the discrete transfer function which is being used to analyze the stability of the model. This transfer function provides us the range of PID parameters for which the model response is stable, given a fixed ratio of the sampling time step to the fundamental vibration period. Current work focuses on optimizing these parameters to minimize an appropriate objective function. We will look for correlation between transfer function predictions and system response predicted by difference equations. We hope to correlate model predictions to experimental results obtained for the system described in the first paragraph.

This work is supported by NASA EPSCoR Grant NCCW-0058.

# DEVELOPMENT OF PIEZOELECTRIC CERAMICS FOR, AND MECHANICAL MODELING OF TRAVELING WAVE MOTORS

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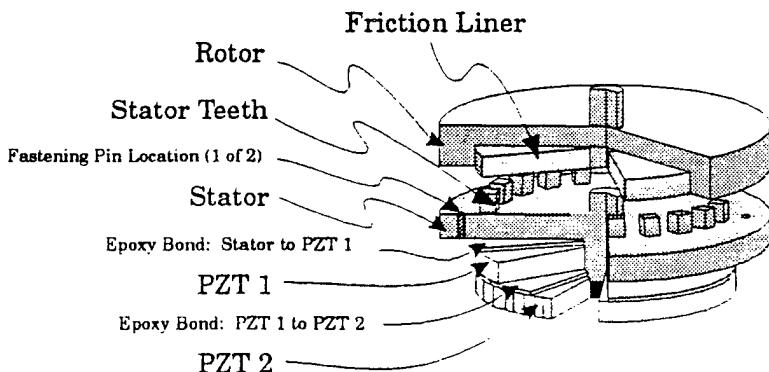
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This paper summarizes findings of a joint program between UMR and Allied Signal to develop traveling wave motors (TWMs). Research efforts are focused on issues related to the piezoelectric elements, analytical modeling of the TWMS, and on overall miniaturization of the motor.

Studies on the piezoelectric have been directed towards optimization in terms of its processing (microstructure, strength, surface finish), electrical properties, and resistance to depoling. In this program we are investigating the effect of these issues using both hard and soft PZT formulations. Motor elements have been prepared using tape cast structures, with emphasis on achieving requisite geometries (down to 50  $\mu\text{m}$  thick) without any machining.

An analytical model was also created to help develop a motor with improved performance and durability. The model allows for the selection of many different material properties and design geometries for better performance and reliability, while accounting for manufacturing limitations such as piezoelectric plate thickness and bond stiffness. An analytical model was also developed for the friction force and stick-slip parameters based on the geometry of the motor and the deformations of the stator by combining theories of plate mechanics with those in contact mechanics. This allows the determination of the torque and rotational speed of the motor, and provides a method for finding the effect of using different contact materials in the contact region.



*Cut-out view of traveling wave motor – element size exaggerated.*

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## HIGH PRECISION PIEZOELECTRIC LINEAR MOTORS FOR OPERATIONS AT CRYOGENIC TEMPERATURES AND VACUUM

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The Jet Propulsion Laboratory evaluated the use of an electromechanical device for optically positioning a mirror system during the pre-project phase of the Pluto-Fast -Flyby (PFF) mission. The device under consideration was a piezoelectric driven linear motor functionally dependent upon a time varying electric field which induces displacements ranging from submicrons to millimeters with positioning accuracy within nanometers. Using a control package, the mirror system provides image motion compensation and mosaicking capabilities. While this device offers unique advantages, there were concerns pertaining to its operational capabilities for the PFF mission. These issues include irradiation effects and thermal concerns. A literature study indicated that irradiation effects will not significantly impact the linear motors operational characteristics. On the other hand, thermal concerns necessitated an in depth study.

To address the thermal issue, we constructed an exact electro-elastic-thermal analytical solution, a finite element model and conducted experimental tests to evaluate the operation of the linear motor at cryogenic temperatures. This study indicated that severe problems arise when operating this device at low temperatures related to thermal mismatches in the materials causing the motor to "lock up" and degradation of the strain coefficients causing a loss in the motor's efficiency. To address these issues, we conducted a parametric study to investigate the impact of geometrical changes and material substitutions on the thermal response of the linear motor. This study indicated that the thermal mismatch problem could be overcome with several possible reconfigurations. We also evaluated the response of the motor's drive element (PZT-5h) at temperatures down to 157 Kelvin. Experimental results indicate that mechanical limitations of the piezoelectric ceramic are strain dependent and electric field independent. Thus, degradation in the piezoelectric strain coefficients at cryogenic temperatures are easily overcome with appropriate modifications to the applied voltage. These preliminary results suggest that the appropriate state variable for modeling/predicting nonlinear and long term response of solid state motors containing piezoelectric material could be strain. Experimental tests on an augmented linear motor at 157 Kelvin demonstrated both clamping and elongation/contraction capabilities supporting the analytical results. Therefore, both analytical and experimental evidence has led to the conclusion that an augmented linear motor can intelligently engineered to operate at the temperature levels of the PFF mission.

Acknowledgments: This program was performed under a California Institute of Technology contract with the National Aeronautics and Space Administration (NASA). The authors would like to thank Burleigh Instruments, Inc. Fishers, NY for graciously donating linear motors to this project.

ACOUSTIC NDE: 1. RESIDUAL STRESS MEASUREMENTS IN PLASTICS,  
2. NON-INVASIVE CURE MONITORING OF COMPOSITES

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1. Longitudinal waves at the critical incidence angle ( $L_{-cr}$ ) have the largest acoustoelastic effect, and thus were utilized to measure residual stress in plastics and glass used for aircraft transparencies. A novel transducer system was developed that greatly facilitates the precise delay time measurement of acoustic waves, by separate crystals receiving reference and leaky wave signals. Absence of water immersion allowed dry contact scanning suitable for field measurements. Simple and low cost systems were developed for stress measurement and flaw detection of laminated plastic structures. For stress measurements in small regions, a spectral technique that allowed measurement of very small phase changes, was demonstrated with acoustic microscopy. A set of transducers and two prototype measurement systems, one analogue and the other a PC based digital, were built. The stress measurements were made on an F-16 canopy.

2. The acoustic wave techniques have been well investigated for cure monitoring due to their ability to directly measure the mechanical properties. However, most of the current sensors are invasive since they need to be inserted into the parts being monitored. Thus the sensors have a limitation in that they can only be placed in the trimmings. Instead, the sensors developed under this project are surface mounted so they hug the part being monitored and are designed so that they can be conveniently housed in the tool. Two different types of sensors were developed, designed, and fabricated and their cure monitoring ability was demonstrated. Lamb wave sensors were made out of a thin stainless steel sheet by sputter depositing interdigital finger patterns using micro photo-lithography. A novel impedance sensor was devised by a rectangular plate of  $\text{LiNbO}_3$  that vibrates in lateral modes with a frequency of a few hundred kHz. Cure tests were conducted on laboratory grade composite samples made by soaking carbon fibers in epoxy.

Acknowledgement: The stress measurement work was supported under SBIR from the WPAT/AFB and the cure monitoring work by NAWADC.

## THE FERROELECTRIC BEHAVIOR AND PIEZOELECTRIC PROPERTIES OF NYLON 5,7 COPOLYMERS

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Following studies of the ferroelectric and piezoelectric properties of the odd-numbered nylon series, nylon 11, nylon 9, nylon 7 and nylon 5, the ferroelectric switching behavior and piezoelectric response of an odd-odd nylon, nylon 5,7, are investigated. Melt-quenched and cold-drawn nylon 5,7 films exhibit typical ferroelectric *D-E* hysteresis behavior when poled under the application of a cyclic electric field at room temperature. The remanent polarization and coercive field of nylon 5,7 are 100 mC/m<sup>2</sup> and 75~80 MV/m, respectively, which are in a good agreement with the previously observed linear relationship between the measured remanent polarization and dipole density for the odd-numbered nylons. In addition, the piezoelectric strain coefficient,  $d_{31}$ , and stress coefficient,  $e_{31}$ , of poled-annealed nylon 5,7 films can also be stabilized up to a temperature of 200°C, with a stable high temperature value of  $d_{31} = 16$  pC/N and  $e_{31} = 23$  mC/m<sup>2</sup>. Melt-quenched  $\beta$ -methylated nylon 5,7 films were also studied. They are found to exhibit no *D-E* hysteresis behavior. Wide-angle X-ray diffraction studies of both unpoled and poled nylon 5,7 films will be presented and discussed along with an interpretation of the switching behavior observed.

This work is supported by ONR and CAFT.

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# ELECTROSTATICS AT ROUGH INTERFACES

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The response of electrostrictive and piezoelectric materials depends on the electric field within the material as well as at material interfaces. Interfacial roughness can significantly influence the field near the interface. In this poster we present results of calculations of the electric field at the interface between a dielectric material and a conducting body.

We consider the electrostatic field in a dielectric material bounded by two, nominally planar, conducting electrodes with small surface roughness. The electrostatic potential is determined analytically in the limit of small roughness using a double perturbation scheme. Results are compared to numerical calculations valid for arbitrary magnitude roughness.

We find that the electric field strength near surface asperities can be much larger than the apparent field strength. In the limit of small height and small width asperities, the field enhancement at the electrode surface becomes independent of the electrode gap width. We also determine the surface-average field enhancement and Maxwell stress in terms of roughness parameters.

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THE COMPLIANCE COEFFICIENTS OF AN AMP (FORMERLY PENNWALT) PVDF  
COPOLYMER

At the Office of Naval Research Transducer Materials and Transducer Workshop in 1992, Acoustical Research and Applications presented measurements of the compliance coefficients of a PVDF copolymer manufactured by Pennwalt. The complex tensile coefficients - the off-diagonal tensile coefficients were measured in a collaborative effort with Drs. Hong Wang and Qiming Zhang of The Materials Research Laboratory - appeared to be reasonable in both magnitude and loss tangent; however, the shear coefficients were about 20 times larger than expected. This paper presents the results of recent measurements made with an updated version of the Dynamic Compliance Apparatus (DCA), points out limitations of the original apparatus, and of the simple model used in its design. (Design and development of the original DCA and the earlier measurements were sponsored by NAVSEA under an SBIR contract monitored by The Naval Undersea Warfare Center Detachment at New London, CT.)

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## DUAL BEAM VIBROMETRY MEASUREMENTS OF ELECTROACTIVE POLYMER FILMS

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The measurement of piezoelectric or electrostrictive strains in thin (~ 1mil) low modulus polymer films requires a non-contact experimental method. To this end, we have developed a dual beam laser Doppler vibrometer and the measurement techniques for obtaining the electromechanical coupling constant of such films. Discussion will emphasize the measurement technique which includes sample geometry, mounting and processing of the signal of the electrically active mode.

In thin films, we measure a strain which is a superposition of the electroactive mode and a flexural mode. Each of these modes has to be extracted from the optical signal from each film surface to obtain the true strain of the electroactive mode, especially for low modulus polymer films. This is in contrast to processing the difference of optical signals from each film surface which we show can overestimate the electroactive strain by an order of magnitude. Examples will be given for the piezoelectric polymer PVDF and an electrostrictive polyurethane.

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6 April 1995

***ABSTRACTS***  
***(continued)***

## PHOTOSTRICTIVE EFFECT IN PLZT CERAMICS AND ITS APPLICATIONS

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Photoacoustic devices, "photophones," which can convert the illuminated light energy directly into the sound energy, will be very significant interfaces to human in the next optical communication age, replacing "telephones" in the current electrical communication age. We have developed a completely new-principle device, using photostrictive materials.

The photostrictive effect is an intriguing phenomenon which exhibits material deformation by the application of light (not due to the thermal expansion!), and is observed in particular piezoelectric materials. Although the mechanism has not as yet been clarified, this is understood as a coupled effect of piezoelectricity with the photovoltaic effect, where under the illumination of light an electric field of the order of several kV/cm is produced.

We have improved the responsivity of the photostriction through the investigations on the ceramic compositions, dopants, and preparation methods. The material used for this application was a PLZT ceramic doped with slight amount of WO<sub>3</sub>. When this ceramic was illuminated with purple-color light (wavelength of 366 nm with the intensity 1 mW/cm<sup>2</sup>), the sample was elongated by 0.01% in size in 1 sec. This small deformation was amplified with a bimorph structure up to 150  $\mu$ m.

The photostrictive bimorph was driven by an alternate irradiation of purple-color light on the two sides around its mechanical resonance frequency of 80 Hz. The tip vibration displacement was remarkably enhanced at about 80 Hz of the driving irradiation frequency. This photo-induced mechanical resonance in the audible frequency range indicates the feasibility of the newly developed photostrictive actuators to future "photophone" applications.

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## FIELD-INDUCED STRAIN AND POLARIZATION SWITCHING MECHANISMS IN PLZT CERAMICS NEAR THE PZT MORPHOTROPIC PHASE BOUNDARY

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Dielectric properties and electric field-induced strain of  $Pb_{1-x}La_x(Zr_yTi_{1-y})O_3$  (abbreviated as PLZT  $100x/100y/100(1-y)$ ) ceramics have been investigated in the compositional range near the PZT rhombohedral-tetragonal morphotropic phase boundary, for La-contents between 0 and 7 at. % and Zr/Ti ratio between 55/45 and 65/35. Domain morphologies from transmission electron microscopy (TEM) were also examined, in order to provide some insights into the polarization switching mechanisms. Common trends in the domain structure with increasing La-content were observed, including: a micron-sized domain structure, a subdomain tweed-like structure, and a polar nanodomain structure. The dominant electromechanical coupling mechanism changed from piezoelectricity for the micron-sized domain structure to electrostriction for the polar nanodomain structure. Compositions along the morphotropic phase boundary in the PLZT phase diagram were found to have relatively high strain, probably resulting from the ease of domain switching. The composition having maximum field-induced strain was PLZT 5/5644. The correspondent polarization switching is suggested to be by a subdomain tweed-like structure.

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Transverse Piezoelectric Mode Piezoceramic Polymer Composites with  
High Hydrostatic Piezoelectric Responses

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In addition to the connectivity of the constituents in a composite, the operation mode also plays an important role in determining the performance of the composite. In this talk, we will present two types of piezoceramic polymer composites developed recently at MRL of Penn State: a 2-2 piezocomposite operated at the transverse piezoelectric (TP) mode and a TP mode honeycomb composite. Both composites exhibit exceptionally high hydrostatic piezoelectric response, high reliability, as demonstrated by the experimental results on these new composites and analytical modeling. Based on analytical models, the optimum design of these composites is also analyzed. One advantage of a TP 2-2 composite, in addition to the high hydrostatic piezoelectric response, is the low fabrication cost. While for a TP mode honeycomb composite, due to the fact that the piezoelectric responses from the three orthogonal directions add together when the transducer is subjected to a hydrostatic pressure, a unique feature of this composite, it has a piezoelectric hydrostatic response considerably higher than those of most other piezoceramic polymer composites.

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# HOMOGENIZATION THEORY FOR PIEZOCOMPOSITES AND APPLICATIONS TO ACOUSTIC TRANSDUCERS

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**ABSTRACT.** The use of materials with negative Poisson's ratio in piezocomposites was proposed in [1] for the large enhancement of hydrophone performance.

We developed an analytical approach (based on homogenization theory) for calculating structural property relationships for materials with degenerating and negative Poisson's ratio [2-4]. Several examples of such composites will be presented.

Applications of homogenization theory to dynamical problems in piezoceramic/polymer composites will be also discussed.

W.A. Smith, *"Optimizing electromechanical coupling in piezocomposites using polymers with negative Poisson's ratio,"* Proc. IEEE 1991 Ultrasonics Symp., IEEE (1991), 661-666.  
Berlyand, L.V., Kozlov, S.M., *Asymptotics for Homogenized Moduli for Elastii Chess-Board Composite*, Archive for Rational Mechanics and Analysis 118 (1992), 95-112.  
Berlyand, L.V., Gendelman, O.V., *Streched checkerboard model of an orthoelastic composite of two vastly different materials*, Abstracts of the Am. Math. Society 14 #3 (1993).  
Berlyand, L.V., Promislow, K.S., *Effective Elastic moduli of a Soft Medium with Hard Polygonal Inclusions and Extremal Behavior of Effective Poissons Ratio*, (submitted for publication).

# COMPUTER SIMULATION OF DOMAIN STRUCTURES AND THEIR KINETICS IN FERROELECTRICS

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A two-dimensional discrete model has been constructed for the study of domain pattern formation in ferroelectrics using molecular dynamics. The model includes both isotropic and anisotropic nonlinear local potential, nearest neighbor interaction and the dipole-dipole interactions.

The nonlinear local potential leads to four degenerate states in the ferroelectric phase of a two dimensional system with square symmetry high temperature phase. However, the nonlocal coupling in the form of nearest neighbor interactions forces the system to form a single domain structure without external constraints. The process of domain pattern formation depends strongly on the cooling rate. Faster quenching of temperature often produces metastable domain configurations with relatively small domain sizes and sometimes even charged domain walls, whereas slow cooling may bring the system to the single domain state or at least produces large domain sizes.

The dipole-dipole interaction can be very important if the depolarization field is not screened. Our simulation results show that the dipole-dipole interaction is in favor of an antiferroelectric state. In a real system the nonlocal coupling which is in favor of single domain state provides a balance to the dipole-dipole interaction, hence, periodic domain patterns can be produced. A uniaxial ferroelectric system is studied as an example using the current model. Both domain walls and antiphase boundaries are produced. The final stable periodic domain configuration can be obtained through extensive annealing processes. The effects of boundary conditions and system size influence will also be discussed.

<sup>†</sup> Research was sponsored by NSF Grant # DMR-92-23847 and ONR Grant # N00014-92-0340.

## SIZE EFFECTS IN FERROIC SOLIDS

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This NSF-MRG program on Size Effects in Ferroelectric Solids involves faculty and students from five different academic programs. The engineering objective is to carry out the basic research required to miniaturize and integrate transducers, capacitors, and other ferroelectric components. The scientific objective is to measure and understand the many interesting scaling phenomena observed in ferroic solids as the size drops below 1  $\mu\text{m}$ . Studies are being carried out on both primary (ferroelectrics, ferroelastics, and ferromagnetics) and secondary ferroics (ferrobielastics, ferroelastoelectrics, etc.) using fine-grained ceramics, small particles, and thin films. In regard to scaling effects, four important size ranges are observed: (1) Large grains (typically 1  $\mu\text{m}$  or larger) contain many domains, and the physical properties are strongly influenced by domain wall motion; (2) smaller grains (around 0.1  $\mu\text{m}$  in size) are often single domain with a very different switching process; (3) at still smaller sizes, the Curie temperature changes and the phase transformation broadens to give spin glass behavior, and finally, (4) as the grain size approaches the atomic scale, each small cluster of atoms behaves independently giving normal paraelectric or paramagnetic behavior. Stages (2) and (3) are the focus for this program, with typical dimensions of 10-100nm.

Six graduate students are employed on the MRG/NSF project. The thesis topics are:

- "Critical Size Phenomena in  $\text{BaTiO}_3$  Thin Films" (J. P. Maria)
- "Size Effects in Glass-Ceramics Containing Ferroic Particles" (D. McCauley)
- "Microwave Resonance Phenomena in Small Ferroic Particles" (M. McNeal)
- "Electrostrictive Properties of Ferroic Nanocomposites" (V. Sundar)
- "Modeling of Domain Wall Motion in Small Ferroic Crystallites" (D. Yang)
- "Critical Size Effects in Sol-Gel Films of Antiferroelectric  $\text{PbZrO}_3$ " (C. Gaskey)

In addition to advising graduate students, the faculty members associated with the MRG project are carrying out research on ferroic materials using spectroscopic ellipsometry (S. Trolier-McKinstry), ceramic processing techniques (T. Shroud), x-ray diffraction and CCD microscopy (K. Uchino), microwave microstrip measurements (S. Jang and J. Fielding), computer simulation of domain wall motions (W. Cao), Landau theory (J. Banavar) and transmission electron microscopy (C. Randall). A brief summary of the objectives of this program will be presented.

# 1996 ONR Transducer Materials and Transducers Workshop

## PRELIMINARY AGENDA

25-27 March 1996

### Sunday, 24 March 1996

7:00-9:00 p.m. Registration – Scanticon Conference Center

### Monday, 25 March 1995

7:30-8:00 a.m. Registration

8:00-8:10 a.m. "Navy Overview"  
Wallace A. Smith and Scott Littlefield (Office of Naval Research)

8:15-8:55 a.m. "Performance Limits in Sensors"  
Thomas B. Gabrielson (NAWC Aircraft Division–Warminster)

9:00-9:30 a.m. "Transducer Studies in the MRL: Overview"  
L. Eric Cross (Materials Research Laboratory, Penn State)

9:35-10:10 a.m. **–Break–**

10:15-10:35 a.m. "Micromachined Ultrasonic Transducers (MUTs)"  
B.T. Khuri-Yakub and I. Ladabaum (Stanford University)

10:40-11:00 a.m. "Monolithic PZT-on-Silicon Monomorph Arrays for Acoustic Imaging"  
J. Bernstein, K. Houston, and L. Niles (C.S. Draper Laboratory); H.D. Chen, K. Li, K.R. Udayakumar, and L.E. Cross (Penn State University)

11:05-11:25 a.m. "Directional Micromachined Accelerometers for Underwater Applications"  
Patrick J. Kelly (NAWC Aircraft Division–Warminster)

11:30-12:13 p.m. *Three Minute Poster Summaries (Group A)*

1. "Two-Dimensional Array Fabrication Using Multilayer Flexible Circuit Technology"  
R.E. Davidsen, S.W. Smith, E.D. Light, and R.B. Ash (Duke University)
2. "Signal to Noise Ratio of 2-D Arrays Using Multilayer PZT"  
C.D. Emery and S.W. Smith (Duke University)
3. "Physical Vapor Deposition of Lead Zirconate Titanate Films for Use in Two-Dimensional, Multi-Layer Ultrasonic Transducer Arrays"  
Robert Kline-Schoder and Shinzo Onishi (Creare Incorporated)
4. "Real-Time 3D Ultrasonic Imaging with a High Density Composite Piezoelectric 2D Array"  
K.R. Erikson, A.M. Nicoli, and T.E. White (Loral Infrared & Imaging Systems Inc.)
5. "High Frequency Ultrasonic Transducers Using Relaxor and Single Crystal Ferroelectrics"  
P.D. Lopath, K.K. Shung, Seung-Eek Park, and T.R. Shrout (Penn State University)

11:30-12:13p.m. *Three Minute Poster Summaries (Group A)–continued*

6. "High-Frequency Piezoelectric Properties of Fine-Grained PZT"  
M.J. Zippardo, K.K. Shung, Wesley Hackenberger, and T.R. Shrout (Penn State University)
7. "Ultrafine Scale Piezoelectric Composite Materials for High Frequency Applications"  
B.G. Pazol, L.J. Bowen, R.L. Gentilman, H.T. Pham-Nguyen, and W.J. Serwatka  
(Materials Systems Inc.)
8. "The Dynamic Behavior of Piezo-composite and its Correlation to the Performance of Ultrasound Transducers"  
J.R. Yuan, K. Liang, P. Marsh, and H.A. Kunkel (ATL Echo Ultrasound)
9. "Resonant Modes, Acoustic Impedance, and Surface Vibration Profiles of Periodic Piezoceramic-Polymer Composite Plates"  
Q.M. Zhang, Xuecang Geng, and Jian Yuan (Penn State University)
10. "Finite Element Study of Varied Impedance Matching Layer Properties"  
Mark R. Draheim and Wenwu Cao (Penn State University)
11. "Piezoelectric Polymer Applications in Solid-State Intracoronary Ultrasound (ICUS) Heart Catheter"  
JoAnne Moody, Michael Eberle, and Dough Stephens (EndoSonics)

12:13 p.m. *Conferee Lunch–Scanticon Conference Center (Deans Hall)*

1:30-1:50 p.m. "Active Control of Structurally Radiated Sound Using an Integrated Piezoelectric Double Amplifier Skin"  
C.R. Fuller, M. Wenzel, and C. Guigou (Virginia Polytechnic Institute & State University);  
B. Xu, Q. Zhang, V. Kugel, and L.E. Cross (Penn State University)

1:55-2:15 p.m. "Development of Transversely Aligned Piezoelectric Fiber Composites for Active Structural Acoustic Control"  
N.W. Hagood, A.A. Bent, and J.P. Rodgers (MIT)

2:20-2:40 p.m. "Monolithic Accelerometer for Smart Actuator Applications"  
Robert D. Corsaro, Joseph D. Klunder, and Brian Houston (Naval Research Laboratory)

2:45-3:05 p.m. "A Concurrently Engineering Adaptive Composite Panel"  
G.H. Koopman, K.L. Koudela, and W. Chen (Penn State University)

3:10-3:50 p.m. **–Break–**

3:50-4:23 p.m. *Three Minute Poster Summaries (Group A)*

12. "Processing Control During the Fabrication of PMN Powders by a Liquid-mix Process"  
Wayne Huebner and Chen-Lung Fan (University of Missouri-Rolla)
13. "A Comparative Study of the Kinetics of Hydrothermally Derived Perovskite-Type Materials"  
B.L. Gersten, M.M. Lencka, and R.E. Riman (Rutgers University)
14. "Novel Chemical Processes for Manufacturing Fine-Scale Piezoelectric Materials"  
W.J. Dawson, J.G. Darab, S.L. Swartz, and D.A. Beckholt (NexTech Materials, Ltd.)

3:50-4:23 p.m. *Three Minute Poster Summaries (Group A)-continued*

15. "Processing and Structure-Property Relationships for Fine Grained PZT Ceramics"  
T.R. Shrout, C.A. Randall, N. K, W. Cao, and W.S. Hackenberger (Penn State University)
16. "Sinter-Forging of Fine Grained Dielectrics"  
G. Risch and T. Shrout (Penn State University)
17. "Low Temperature Processing of Lead Zirconate Titanate Piezoelectric Glass-Ceramics"  
B. Houng, C.Y. Kim, Y.D. Kim, and M.J. Haun (Colorado School of Mines)
18. "Intelligent Process Models for Better Synthesis Processes for Ferroelectrics"  
R.E. Riman, M.M. Lencka, I. Petrovic, and E.A. Gulliver (Rutgers University)
19. "Synthesis of Modified PbTiO<sub>3</sub> Powders and Fibers by a Liquid-mix Process"  
Wayne Huebner and Chen-Lung Fan (University of Missouri-Rolla)
20. "Tape Casting of Powder Materials Using Intelligent Process Control Methods"  
M.R. Pascucci, J.J. Bausch, III, and R.N. Katz (Worcester Polytechnic Institute)
21. "A Novel Approach to Fabricating High Q<sub>m</sub> PZT"  
J. Alexander Chediak and Steven M. Pilgrim (Alfred University)
22. "Studies on the Electromechanical Properties of PbTiO<sub>3</sub>-Based Ceramics"  
W. Huebner and W.R. Xue (University of Missouri-Rolla)

4:23-6:00 p.m. Poster Viewing ('Group A' authors please be at your posters)

**Reception-Scanticon Conference Center (Presidents II and pre-function area)**

6:30 p.m. **Cash Bar**  
7:00-9:00 p.m. **Reception and Poster Viewing**

**Tuesday, 26 March 1996**

7:30-8:00 a.m. -Coffee-

8:00-8:40 a.m. "A 128 × 4 Channels 1.5D Curved Linear Array for Medical Imaging"  
P. Tournois, S. Calisti, and Y. Doisy (Thomson Microsonics);  
J.M. Bureau and F. Bernard (Thomson CSF-LCR)

8:45-9:05 a.m. "Phase Aberration Correction in Two Dimensions Using a Deformable Array Transducer"  
L.L. Ries and S.W. Smith (Duke University)

9:10-9:40 a.m. "Dynamic Imaging of Piezocomposite Transducers"  
I. Perez (NAWC Aircraft Division-Patuxent River); M. Ryan (NAVMAR Applied Science Corp-Warminster); R. Gentilman, D. Fiore, and L. Bowen (Materials Systems Inc); J. Yuan (Echo Ultrasound); and W. Cao (Penn State University)

9:45-10:15 a.m. -Break-

10:20-10:40 a.m. "Finite Element Modeling of a 2-Dimensional Transducer Array"  
C. Desilets (UltraSound Solutions); D.K. Vaughan, N. Abboud, G.L. Wojcik, and J. Mould, Jr. (Weidlinger Associates)

10:45-11:05 a.m. "Nonlinear Modeling Issues for Ultrasound Tranducers"  
G.L. Wojcik, J. Mould, Jr., D. Vaughan, and N. Abboud (Weidlinger Associates)

11:10-11:30 a.m. "Performance Analysis of a Low-Frequency Barrel-Stave Flextensional Projector"  
D.F. Jones (Defence Research Establishment Atlantic); N.N. Abboud, G.L. Wojcik, and D.K. Vaughn (Weidlinger Associates)

11:35-12:05 p.m. *Three minute poster summaries (Group B)*

23. "Development of Ultra-Fine Scale Piezoelectric Fibers for Use in High Frequency 1-3 Transducers"  
"R.J. Meyer, Jr., S. Yoshikawa, and T.R. Shrout (Penn State University)

24. "Forming and Sizing of Fine Pb(Zr,Ti)O<sub>3</sub> Fibers"  
J.D. French, G.E. Weitz, J.E. Luke, R.B. Cass (Advanced Cerametrics Incorporated);  
A. Safari, V.F. Janas, and B. Jadidian (Rutgers University)

25 (I). "PZT Microrod Composite Ultrasonic Transducers"  
M.T. Strauss, M.V. Parish, and D. Ouellette (CeraNova Corporation)

26. "1-3 PZT Composite Transducer Research at ARL/PSU"  
W.J. Hughes (Penn State University)

27(I). "Determining Material Constants, Losses, Frequency Dispersion, and Non-Linearity with the Piezoelectric Resonance Analysis Program (PRAP)"  
R. Tasker (TASI Technical Software)

28. "Finite Element Study on Random Piezocomposite Transducers"  
Wenkang Qi, and Wenwu Cao (Penn State University)

29. "Closing the Loop in Transducer Modeling - Materials, Structures and Acoustic Field"  
Wenwu Cao (Penn State University)

30. "An Alternative Equivalent Circuit for the Unloaded Piezoelectric Resonator"  
S. Sherrit, H.D. Wiederick, B.K. Mukherjee (Royal Military College of Canada);  
and M. Sayer (Queen's University)

31. "Effect of Mechanical Stress on the Electromechanical Performance of PZT Ceramics"  
Jianzhong Zhao, Q.M. Zhang, and L.E. Cross (Penn State University)

32. "State of the Art Piezoelectric and Optical Components"  
Russell S. Petrucci (Valpey-Fisher Corporation)

12:00 noon *Conferee Lunch – Scanticon Conference Center (Deans Hall)*

1:30-1:50 p.m. "Recent Advances in Piezocomposite Materials, Transducers, and Arrays at MSI"  
R. Gentilman, D. Fiore, H. Pham-Nguyen, W. Serwatka, B. Pazol, C. Near, P. McGuire, and L. Bowen (Materials Systems Inc.)

1:55-2:15 p.m. "Constant Beamwidth 1-3 Composite Tranducer"  
C.W. Allen and W.J. Hughes (Penn State University)

2:20-2:40 p.m. "A Sonar Application of 1-3 Piezocomposite Material"  
F. Geil (Northrup-Grumman Oceanic Systems, Annapolis, MD); K. Webman (Naval Undersea Warfare Center-New London); R.Ting and M. Pecoraro (Naval Undersea Warfare Center-Orlando); R. Gentilman and W. Serwatka (Materials Systems Inc.)

2:45-3:05 p.m. "Processing of Fine-Scale PZT Fiber and Fiber/Polymer Composites"  
A. Safari, V.F. Janas, R.P. Schaeffer, B. Jadidian, R.K. Panda, A. Bandyopadhyay, and S.C. Danforth (Rutgers University)

**3:10-3:40 p.m. -Break-**

**3:45-4:18 p.m. Three Minute Poster Summaries (Group B)**

33. "Thin Films of Equiatomic Titanium-Nickel-I: Sputter Deposition, Mechanical Properties, and Applications"  
David S. Grummon and Thomas J. Pence (Michigan University)

34. "Thin Films of Equiatomic Itanium-Nickel-II: Modeling Thermotractive Transformations to Predict Thermomechanical and Hysteresis Response"  
Thomas J. Pence and David S. Grummon (Michigan University)

35. "Design and Fabrication of Piezocomposite Smart Panels for Active Surface Control"  
D. Fiore, R. Gentilman, H. Pham-Nguyen, W. Serwatka, P. McGuire, and L. Bowen (Materials Systems Inc.)

36. "Materials for Integrated Sensor/Actuator Combinations"  
Joseph P. Dougherty (Penn State University)

37. "Antivibration System with Piezoelectric Polymer Actuators"  
H. Schmidt, D. Brandt, D. Rosenberg, and M. Williams (Montana University)

38. "Large Area Piezoelectric Composite Arrays"  
W.A. Schulze and M.J. Creedon (Alfred University)

39(I). "Production of Distorted 3-3 Hydrophone Composites from Reticulated Ceramics"  
D.A. Norris, T.B. Sweeting, L.A. Strom, and R.M. Utt (Hi-Tech Ceramics)

40. "Electrostriction in Polyurethanes-Morphology Dependence"  
E. Balizer (Naval Surface Warfare Center-Silver Spring); F. Guillot and J. Jarzynski (Georgia Institute of Technology); and J.D. Lee (Naval Surface Warfare Center-Silver Spring)

41. "Mesoscopic Instability in Terfenol-D Films"  
Manfred Wuttig (University of Maryland)

42. "Magnetostriction of Terfenol-D Single Crystals"  
A.E. Clark (Clark Associates); M. Wun-Fogle and J.B. Restorff (Naval Surface Warfare Center-Silver Spring)

3:45-4:18 p.m. *Three Minute Poster Summaries (Group B)–continued*

43. "Improved Piezoelectric Ceramic-Polymer Composites for Hydrophones Applications"  
C.Cui, R.H. Baughman, Z. Iqbal, T.R. Kazmar, and D.K. Dahlstrom (Allied Signal)

4:18-6:00 p.m. Poster Viewing ('Group B' authors please be at your posters)

**Banquet–Scanticon Conference Center (Deans Hall)**

6:30 p.m.—Cash Bar

7:00 p.m.—Dinner

8:30-10:00 p.m.—Poster Viewing (Presidents II)

**Wednesday, 27 March 1996**

7:30-8:00 a.m. Coffee

8:00-8:20 a.m. "Piezoelectric Ceramics From the Rostov State University, Russia"  
Manfred Kahn, Steve Sullivan (Naval Research Laboratory); and Mark Chase  
(Potomac Research Incorporated)

8:25-8:45 a.m. "Field Induced Antiferroelectric-to-Ferroelectric PLZST Ceramics"  
S. Yoshikawa, S.E. Park, K. Markowski, M.J. Pan, T. Shrout, and L.E. Cross  
(Penn State University)

8:50-9:20 a.m. *Three Minute Poster Summaries (Group C)*

44. "Domain Related Phase Transitions in Lead Zinc Niobate Relaxor Ferroelectric Single Crystals"  
Maureen L. Mulvihill, L. Eric Cross, Kenji Uchino, and Wenwu Cao (Penn State University)

45. "Crystal Growth and Ferroelectric Related Properties of  $(1-x) \text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3 - x \text{PbTiO}_3$ "  
Seung-Eek Park, Maureen Mulvihill, George Risch, Mike Zippardo, and Thomas R. Shrout  
(Penn State University)

46. "Antiferroelectric-to-Ferroelectric Phase Switching PLZST Ceramics-I. Structure, Compositional  
Modification and Electric Properties"  
S.E. Park, K. Markowski, S. Yoshikawa, and M.J. Pan (Penn State University)

47. "Antiferroelectric-to-Ferroelectric Phase Switching PLZTS Ceramics-II. The Effect of Pre-Stress  
Conditions on the Strain Behavior"  
M.J. Pan, S.E. Park, K. Markowski, and S. Yoshikawa (Penn State University)

48. "In-situ x-ray diffraction study of the antiferroelectric-ferroelectric phase transition in PLSnZT"  
C.T. Blue and J.C. Hicks (NCCOSC/RDT&E Division); S.E. Park, S. Yoshikawa, and L.E. Cross  
(Penn State University)

49. "The Effect of Annealing Temperature on the Formation of  $\text{SrBi}_2\text{Ta}_2\text{O}_9$  (SBT) Thin Films"  
D. Ravichandran, K. Yamakawa, R. Roy, A.S. Bhalla, S. Trolier-McKinstry, R. Guo, and L.E. Cross  
(Penn State University)

50. "Thin Film Actuator Materials"  
S. Trolier-McKinstry, K. Yamakawa, J. Lacey, J. Shepard, T. Su, and F. Xu (Penn State University)

8:50-9:20 a.m. *Three Minute Poster Summaries (Group C)–continued*

51. "Molecular Dynamics Simulations of PMN Ceramics"  
G. Kavarnos (NUWC-New London)
52. "Thermochemistry and Non-Ohmic Electrical Contacts in Electro-Ceramic Materials"  
Clive Randall and Dave Cann (Penn State University)
53. "The Role of Polarization Mechanisms in Electrostrictive Effects for Low Permittivity Glasses and Ceramics"  
V. Sundar, R. Yimnirun, and R.E. Newnham (Penn State University)

**9:20-10:00 a.m. –Break–**

10:00-10:30 a.m. *Three Minute Poster Summaries (Group C)*

- 54 (I). "Studies on the Development of Piezoelectric Traveling Wave Motors"  
W. Huebner, D. Stutts, and J. Friend (University of Missouri-Rolla); and C. Mentesana (Allied Signal)
55. "Compact Ultrasonic Motor"  
Amod Joshi, Seok Jin Yoon, and Kenji Uchino (Penn State University)
56. "Rainbow Actuator Stacks and Arrays"  
Gene Haertling (Clemson University)
57. "Stress and Fatigue Estimation in Multi-Layer Ceramic Actuators Using an Internal Strain Gauge"  
H. Aburatani and K. Uchino (Penn State University)
58. "Cracking in Ferroelectric Ceramic Multilayer Actuators"  
X. Gong, H. Yu, Z. Suo, and R. McMeeking (University of California–Santa Barbara)
59. "Ceramic-Metal Composite Transducers for Underwater Acoustic Applications"  
J.F. Tressler, W. Cao, K. Uchino, and R.E. Newnham (Penn State University)
60. "Bimorph Based Double Amplifier: A Potential Transducer Used in Air Acoustics"  
Baomin Xu, Q.M. Zhang, V.D. Kugel, Qingming Wang, and L.E. Cross (Penn State University)
62. "Bimorph-Based Air Transducer: A Model"  
V. Kugel, Q.M. Zhang, Baomin Xu, Qingming Wang, and L.E. Cross (Penn State University)
63. "The 'Moonie' and 'Cymbal' Electromechanical Actuators"  
A. Dogan, J.F. Fernandez, K. Uchino, and R.E. Newnham (Penn State University)
64. "Results of 8mm Ultrasonic Minimotor Development Using Design of Experiments"  
Anita M. Flynn (University of California–Berkeley)

10:40-12:00 noon Poster Viewing ('Group C' authors please be at their posters)

**12:00 noon Conferee Lunch–Scanticon Conference Center (Deans Hall)**

1:30-1:50 p.m. "Reliability of Ceramic Actuators"  
Kenji Uchino (Penn State University)

1:55-2:15 p.m. "Vibration Modes of Tangentially Poled PZT Hollow Spheres"  
S. Alkoy, A. Dogan, and R.E. Newnham (Penn State University); A.C. Hladky (IEMN-Department I.S.E.N.); and J.K. Cochran (Georgia Institute of Technology)

2:20-2:40 p.m. "Novel Transducer Materials and Fabrication"  
Thomas R. Shrout (Penn State University)

2:45-3:05 p.m. "Quasistatic Measurements of the Converse and Direct Piezoelectric Charge Coefficient in Piezoelectric Ceramics"  
S. Sherrit, R.S. Stimpson, H.D. Wiederick, and B.K. Mukherjee (Royal Military College of Canada)

**3:10-3:40 p.m. -Break-**

3:45-4:05 p.m. "Microscopic Strain Measurements of PMN-Based Electrostrictors"  
Sean P. Leary and Steven M. Pilgrim (Alfred University)

4:05-4:25 p.m. "Characterization of PMN-PT-LA (0.90/0.10/1%) for Use in Sonar Transducers"  
E.A. McLaughlin, J.M. Powers, M.B. Moffett, and R.S. Janus (NUWC-New London)

4:30-4:50 p.m. "Domain-Like Organizations in Ferroelectrics and Antiferroelectrics Containing Quenched Randomness"  
Dwight Viehland (University of Illinois-Urbana)

**4:55 p.m. Adjourn**

## 1996 ONR Transducer Materials and Transducers Workshop - Attendance List

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# **1996 ONR Transducer Materials and Transducers Workshop**

## ***Abstract Booklet***

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***Morning  
Presentations***

## PERFORMANCE LIMITS IN SENSORS

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The quest for improved sensor performance — lower minimum detectable signals, smaller size, lower power — is constant and intense. Consequently, the frequent lack of emphasis on fundamental limits in the initial stages of sensor design is surprising. Development of new transduction technologies is exciting but the cost of ignoring intrinsic self-noise is often frustration and wasted time and effort. Careful noise analysis early in the design process can suggest approaches for better performance; careful measurement of sensor noise can uncover deficiencies in design or construction. In fact, serious examination of sensor noise can often reveal more about the fundamental workings of a sensor than can measurement of its transduction response. The usual assumption that the preamplifier dominates the noise of a sensor system, while sometimes true over limited bands, often leads either to suboptimal performance or to unrealistic expectations. Many other sources of noise such as thermal-equilibrium agitation of mechanical elements, internal Johnson noise, equilibrium and non-equilibrium shot noise, 1/f noise, stress-induced noise in piezoceramics, and various optical noise sources in fiber sensors can set the ultimate performance limits.

An increasingly popular strategy in sensor development is to use microfabrication techniques to reduce sensor size. In translating devices and structures from scales of centimeters to scales of micrometers, effects that were unimportant at the larger scales can become dominant at the smaller scales. This is true even if there is no significant change in the physical behavior of the materials in and around the structures. The problem is compounded when the size is reduced to the point at which the characterization of the materials themselves changes. While microfabrication has made dimensional downscaling to the micrometer level practical, intuition and "engineering principles" developed on the macroscale may lead to unsound devices during that translation. Successful miniaturization of devices and structures depends on an understanding of the changing roles of various forces, energies, and interactions in the smaller scales. Even with this understanding, optimum performance may not be achieved without considering alternate strategies for solving the basic problem. Just because a particular device is very successful at a certain function at one scale does not mean that a scaled down version of that device is the best approach at another scale.

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## TRANSDUCER STUDIES IN MRL: OVERVIEW

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### ABSTRACT

In materials studies the search for perovskite structure solid solutions which encompass morphotropic ferroelectric:ferroelectric phase transitions is continuing. Preparation techniques have been refined to produce the full range of lead indium niobate:lead scandium tantalate (PIN:PST) solid solutions and measurements are proceeding on both ordered and disordered compositions. In lead scandium tantalate:lead titanate, compositions near the MPB have been shown to have excellent properties in polycrystal form and are prime a target now for study in single crystal form.

In the phase switching antiferroelectric:ferroelectric lead tin zirconate titanate (PSnZT) compositions "square loop" switching has been observed in both ceramic and thin film systems. Compositions have been demonstrated in which the switch forward field does not change from 25°C to 75°C, and methods are being explored to close up the hysteresis between forward and backward switching. Volume strains up to 0.6% are encompassed at high fields but more work is needed to sort out the polarization:strain coupling mechanisms.

For the lead zirconate titanate (PZT) system the interest in performance changes under elastic stress has continued with studies of both  $d_{33}$  and  $d_{31}$  as functions of stress and electric field in both hard and soft compositions. Scaling effects are also under study as preparation techniques are refined to produce excellent fine grain ceramics. For grain sizes below 0.7  $\mu$ meter, the separation of 90° walls changes from the accepted scaling and wall interactions on poling are under study. Modeling of domains under different boundary conditions is also being explored using finite element computer modeling.

For lead magnesium niobate, new concerns have emerged regarding practical use under bias and a new study of aging is being accomplished under both electric and elastic bias fields.

In the composite systems, the simplified cymbal design of the flextensional "moonie" type actuator is proving superior in all aspects of performance, both as a sensor and as an actuator. New studies of performance at the Applied Research Laboratory at Penn State have shown most promising initial results. Techniques to electrode the hollow PZT micro-spheres proved very difficult, but new studies using hemispherical outer surface electrodes have shown most interesting resonant spectra.

Work has been continued on a range of transversely poled 2:2 type composite structures to explore the role of face plates and edge caps on performance and to define new and simplified assembly techniques. The additional work has confirmed the advantageous low electrical:low acoustic impedance characteristics of these composites.

For conventional tape case multilayer actuators work has continued on control of microcracking and has evolved a new floating electrode structure which reduces cracking and also permits an alternative mode of acoustic emission testing. In resonant structures new compositions are being studies to reduce heating, and a new type of antiresonant drive examined which also reduces power loss.

A new type of zig-zag composite which permits control of surface motion in two dimensions is being explored and a large parallel program is now examining double amplifier concepts for air acoustic control.

The whole program is underpinned by processing studies which examine new concepts like rate controlled sintering to produce better samples of the broad range of ceramic and single crystal elements essential for the experimental studies.

## MICROMACHINED ULTRASONIC TRANSDUCERS (MUTs)

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Transducers capable of transmitting 1 to 12 MHz ultrasound in air have been reported. It has also been predicted that these novel surface micromachined devices should enable ultrasonic systems with a dynamic range of 100 dB. A deepened understanding of the devices has lead to the construction of the first airborne ultrasound transmission system achieving 100 dB of dynamic range. Results are presented which confirm the 100 dB dynamic range at various frequencies in the MHz range. Experiments depicting the emission of ultrasound into air, continued propagation through a test material, coupling into the air, and airborne detection are discussed. Test materials for through-transmission experiments include glass, Plexiglas, paper, aluminum, and steel. A heuristic treatment of the fabrication and theory of operation of the transducer is summarized. The treatment concludes with a description of efforts to produce immersion transducers with similar technology. This work as been supported by the U.S. Office of Naval Research.

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## MONOLITHIC PZT-ON-SILICON MONOMORPH ARRAYS FOR ACOUSTIC IMAGING

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Arrays of millimeter sized ferroelectric monomorph sonar transducers have been built using sol-gel PZT on micromachined silicon wafers. 8 X 8 monolithic arrays have been tested in water in the frequency range 0.5 to 2 MHz using off-chip multiplexing to address individual elements. Transducer diaphragms were 0.3 to 0.44 mm square monomorphs with 5  $\mu$ m thick PZT on 10  $\mu$ m thick silicon. Improvements to the sol-gel process have yielded high quality crack-free PZT films up to 12 microns in thickness. Greater thickness PZT leads directly to higher sensitivity and figure of merit for acoustic transducers. The longitudinal piezoelectric coefficient  $d_{33}$  is 246 pC/N, a magnitude close to that of a bulk ceramic. Remanent polarization of 28 micro-C/cm<sup>2</sup>, a coercive field of 30 kV/cm, and dielectric constant equal to 1400 were measured on these films. Results of air and in-water testing are presented including frequency response, beam patterns and output capacitance. Potential applications for these transducers are high frequency imaging sonars, medical ultrasound, ultrasonic communication links, and flaw detection (NDT).

System concepts will be presented for a 3-D underwater acoustic imaging system for use in highly turbid waters. While such a system may be implemented by beamforming samples from a sensor array, beamforming in two spatial axes is very computationally intensive, requiring on the order of  $10^{10}$  multiply-add operations per image frame. Instead, a concept for an acoustic lens system will be described with a retinal array of 100 x 100 sensors. A recent test program at Draper developed a four-element acoustic lens for this purpose. A laboratory test system for acoustic image acquisition will be described, followed by test results and acoustic-lens based images. The images are remarkable by acoustics standards, and such a system would be of significant utility to anyone operating in otherwise zero visibility water.

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Abstract

DIRECTIONAL MICROMACHINED ACCELEROMETERS FOR UNDERWATER APPLICATIONS

A substantial amount of emphasis has been placed on the development of micromachined accelerometers as a means to improve the performance of weapons and surveillance systems. The Naval Air Warfare Center Aircraft Division, Warminster (NAWCAD, War) under the sponsorship of the Office of Naval Research (ONR) has been funding the National Aeronautics and Space Administration (NASA)- Jet Propulsion Laboratory (JPL), Division of the California Institute of Technology (CalTech) in Pasadena, California to develop a miniature directional high-sensitivity electron tunneling accelerometer whose noise characteristics will meet the Sea-State Zero noise curve in the 10 Hz to 1000 Hz band. The development of the prototype JPL accelerometer culminated in a successful test in June 1994 at NAWCAD, War of single axis device interfaced through the JPL breadboard electronics. The prototype accelerometer's calibration and noise characteristics met specifications around resonance. Since the prototype test, efforts to fabricate more sensors in a mass production manner and mount them with miniature electronics on a structural frame in an 8 cm<sup>3</sup> titanium sphere have had the typical fabrication and integration issues associated with first article. These issues have led to process improvements during assembly of the accelerometer, mounting of the electronics and a better understanding of the underlying principles and materials properties associated with the operation of the device. This paper will present the test results of the prototype accelerometer, the fabrication and integration issues, the improvements in the accelerometer design and options for improving the accelerometer by materials selection or process mechanism. The latest design is being assembled for integration into several spheres. The design emphasis of the present two year effort is improvements in the acoustic performance below and above resonance, improvement in seismic performance below resonance, improvements using digital control electronics for multiplexing, two axis and three-axis sensors that are free-floating or bottom anchored, and a miniature compass.

**25 March 1996**

***Morning***  
***Poster Presentations***

## TWO-DIMENSIONAL ARRAY FABRICATION USING MULTILAYER FLEXIBLE CIRCUIT TECHNOLOGY

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The fabrication of practical two-dimensional (2-D) transducer arrays is difficult due to the high density of connections which must be made to the individual elements. A typical 2-D array operating at 2.5 MHz has  $40 \times 40 = 1600$  elements, each measuring  $350 \mu\text{m} \times 350 \mu\text{m}$ . Typically, 256 of these elements are electrically connected to the scanner. Previously we have developed 2-D array fabrication techniques involving hand wiring and multilayer thick film ceramic technology. We will describe a new fabrication method which is based on flexible circuit technology. With this technique, the electrical path from the element to the transducer connector is made with multiple layers of metal traces on polyimide film connected with vias. This relatively low cost technique has the advantages of reducing design and fabrication times and improving the acoustic performance of the 2-D array backing.

We have developed a 120 element single layer flex connector for our initial evaluation of this technique. We first drilled  $100 \mu\text{m}$  diameter via holes in a  $75 \mu\text{m}$  thick polyimide substrate. The vias were filled with conductive epoxy to allow a signal path from the elements to the traces on the back side of the polyimide. Then,  $200 \text{ \AA}$  of chrome and  $1,000 \text{ \AA}$  of gold were sputtered on the substrate. The metal was etched using standard photolithographic techniques, leaving the fan-out pattern from the elements to four flex connector inserts. A  $635 \mu\text{m}$  thick piece of Motorola HD3203 material and a silver epoxy matching layer were bonded to the substrate and elements were cut using a dicing saw. Testing of the completed array included vector impedance, pulse-echo sensitivity, and pulse waveform measurement. The completed array was integrated with our scanner and used to make real-time images.

We have recently designed a 256 element multilayer flex connector which has 5 metal layers. Signals are routed on 2 internal trace layers while an internal ground layer is included to reduce electrical crosstalk. The traces are terminated with four double sided 72 position edge connectors. The details of this design will be presented along with our initial results.

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**TITLE: SIGNAL TO NOISE RATIO OF 2-D ARRAYS USING  
MULTILAYER PZT**

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*Abstract* - In our previous work, we examined the improved transmit efficiency of 2-D array transducers using multilayer PZT. In phased array imaging for medical diagnostic ultrasound, the transducer element sensitivity also limits the detection of receive signals. Therefore, the transducer element receive signal to noise ratio (SNR) should be maximized. Typically, a coaxial cable connects the single layer transducer element to a high impedance amplifier in the scanner. However, the electronic noise from the amplifier usually dominates the thermal noise of the transducer which degrades the overall SNR. In this simulation study, a 2-D array element resonant at 3 MHz was connected to a high impedance amplifier. The amplifier was adjacent to the element or connected to the element through a coaxial cable. Three different types of high impedance amplifiers were compared in the study, a bipolar transistor, a MOSFET, and a JFET. Each amplifier had a different voltage noise level and current noise level. The SNR across the input of the amplifier was calculated over the transducer bandwidth. In addition to different amplifier configurations, the number of PZT layers in the 2-D array element was allowed to vary. The results show a single layer element with a MOSFET or JFET located adjacent to the transducer element improves the SNR compared to a bipolar amplifier connected to the element with a coaxial cable by approximately 9 dB. However, similar improvement in SNR can be accomplished by using a five layer PZT element with a coaxial cable connection to a bipolar amplifier.

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**PHYSICAL VAPOR DEPOSITION OF LEAD ZIRCONATE TITANATE  
FILMS FOR USE IN TWO-DIMENSIONAL, MULTI-LAYER  
ULTRASONIC TRANSDUCER ARRAYS**

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Two-dimensional ultrasonic transducer arrays will significantly improve ultrasound image quality because they enable dynamic control of the ultrasound beam in two directions. Two key problems in the manufacture of miniature two-dimensional ultrasonic transducer arrays are the ability to fabricate the small structures and low transducer element sensitivity. The low sensitivity is due to small clamped capacitance and can be increased by employing multi-layer structures (i.e. electrically in parallel and acoustically in series). However, utilizing multi-layer structures only increases the manufacturing difficulties.

We have developed and demonstrated an innovative microfabrication technique to manufacture low-cost, two-dimensional ultrasonic transducer arrays with multi-layer piezoelectric elements for low impedance and high sensitivity. The manufacturing approach is completely scalable to fabrication of transducer arrays in the frequency range of 1-100 MHz in dense or sparse array designs. Our approach employs the following processes for the transducer component fabrication: (1) Physical Vapor Deposition (PVD or sputtering) of high-quality, dense, perovskite lead zirconate titanate (PZT) films as the piezoelectric material using reactive sputtering of a metallic target and (2) the novel use of state-of-the-art photolithography and masking to provide the interlayer electrodes and element interconnections.

To date, we have deposited 42  $\mu\text{m}$  thick PZT films using reactive sputtering. The material possesses the following measured ferroelectric properties: a dielectric constant ( $\epsilon_r$ ) of 695, a dissipation factor ( $\tan \delta$ ) of 0.027, a remnant polarization ( $P_r$ ) of 19  $\mu\text{coul}/\text{cm}^2$ , and a coercive field ( $E_c$ ) of 98 kV/cm. In addition, we have fabricated single layer and multi-layer transducers which have a measured natural frequency of approximately 33 MHz and a measured coupling coefficient of 0.18. As expected, the multi-layer structure exhibits reduced impedance as compared to the single layer transducer structure.

This research has been supported by the Office of Naval Research through the Small Business Innovation Research (SBIR) Program contract number N00014-95-C0364.

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# REAL-TIME 3D ULTRASONIC IMAGING WITH A HIGH DENSITY COMPOSITE PIEZOELECTRIC 2D ARRAY

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Real-time 3D ultrasound imaging has the potential for making dramatic improvements in underwater as well as medical imaging applications. Until recently, fabrication difficulties have limited even the most advanced systems to one-dimensional beamformed arrays, typically using mechanical scanning in the out of plane dimension together with off-line reconstruction, making real-time 3D imaging impossible.

To realize the required number and density of array elements and the interconnection to the associated electronics, LIRIS' has developed a 42 x 64 (2688 total) element 5 MHz 2D array, hybridized to a custom CMOS integrated circuit. Results to date have demonstrated bistatic real-time imaging with three dimensional resolution of 1mm in a tissue equivalent test object.

In this paper, we describe our work-in-progress with a 128 x 128 (16,384 total) element array and 3D real-time imaging system with an emphasis on the array and integrated circuit. Favorable tradeoffs using composite piezoelectric materials, enabled by LIRIS' high-density interconnection technology are discussed. Through massively parallel, focal plane processing, the goal of real-time 3D ultrasonic imaging in the low MHz frequency range is now achievable.

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# HIGH FREQUENCY ULTRASONIC TRANSDUCERS USING RELAXOR AND SINGLE CRYSTAL FERROELECTRICS<sup>†</sup>

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Two novel transducer materials were investigated through both theoretical calculations and experimental design. High frequency single element ultrasonic transducers were constructed using thickness mode single crystal PZN ( $k_t \sim 0.541$ ,  $\epsilon_{33}^s \sim 250$ ), and thickness mode single crystal PMN-PT ( $k_t \sim 0.541$ ,  $\epsilon_{33}^s \sim 820$ ). Two sets of transducers were constructed from each material, one optimized for pulse echo imaging, while the other was air backed to make efficiency measurements. Individual element responses were compared to theoretical results obtained via the one dimensional KLM model, and calculated beam patterns were compared with tomographically reconstructed Schlieren images. In addition, the response of these elements was compared to the response of industry standard PZT elements of the same dimensions. To take advantage of the high observed  $k_{33}$  in PMN (~0.92) and to utilize the capacitance provided by the high  $\epsilon_{33}$ , 2-2 composites of single crystal PMN were constructed using a dice and fill technique. Behavior of these transducer sets were compared with the responses of the monolithic transducers.

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# HIGH-FREQUENCY PIEZOELECTRIC PROPERTIES OF FINE-GRAINED PZT<sup>†</sup>

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The piezoelectric properties of polycrystalline materials have been shown to change with the operating frequency of a device. In order to design transducers which operate at frequencies greater than 0 MHz, it is necessary to accurately measure the high-frequency properties. Properties from 10 to 95 MHz have been evaluated for a number of ceramics, including commercial variations of PZT5-A and PZT5-H, and also new materials designed to have a microstructure consisting of finer grains. Conventional materials were examined under an SEM and found to have grains on the order of 3 to 5  $\mu\text{m}$ , whereas the fine-grained PZT showed average grain sizes from 0.2 to 2.5  $\mu\text{m}$ . The fine-grained materials showed properties comparable to conventional PZT. Measured relative clamped dielectric constants ranged between 550 and 1010 for the fine-grained PZT and between 660 and 1300 for the conventional PZT. Measured thickness coupling coefficients ( $k_t$ ) were also comparable. The material properties of the conventional PZT were found to decrease significantly at frequencies above 20 MHz. The fine-grained materials were found to maintain properties up to about 45 MHz and above that showed less reduction than conventional PZT. At a thickness corresponding to resonance at 95 MHz the fine-grained material showed a  $k_t$  of 0.435 while it was not possible to construct such resonators from the conventional materials due to lack of mechanical integrity.

<sup>†</sup>This work was supported by the Whitaker Center for Medical Ultrasonic Transducer Engineering and the Office of Naval Research.

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## ULTRAFINE SCALE PIEZOELECTRIC COMPOSITE MATERIALS FOR HIGH FREQUENCY APPLICATIONS

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Intravascular and endoscopic acoustic imaging typically require high ultrasonic operating frequencies, over 10 MHz. At these frequencies the ceramic transducer dimensions become very small (on the order of 20 to 80  $\mu\text{m}$ ). These piezoelectric transducer array requirements severely challenge the ability of conventional PZT ceramic technologies in terms of cost-effective fabrication. To meet the demand for such transducer shapes, MSI has developed an injection molding process for rapid mass production of piezoelectric ceramics in complex shapes at low cost.

Using this technique, MSI has produced ultrafine scale 2-2 piezocomposite arrays having PZT elements under 25  $\mu\text{m}$  wide with pitches under 50  $\mu\text{m}$ . This paper reviews recent advances in the capabilities of the injection molding process for fabricating extremely fine element dimensions to net shape and compares the responses of one configuration with conventionally diced arrays.

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## The Dynamic Behavior of Piezo-composite and its Correlation to the Performance of Ultrasound Transducers

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Because the piezo-composites provide a new range of material properties, and can be tailored to specific device requirements, the dynamic behaviors, properties and applications of this material have been extensively studied both theoretically and experimentally. For the most common class of 1-3 piezo-composite, the upper frequency range of operation may be limited by undesirable resonances associated with the periodic dicing structures and composite constituent materials.

The thickness coupling coefficient  $k_t$  for the thickness mode reaches the maximum value when the thickness mode is decoupled from lateral resonant modes. Seeking a way to reduce the undesirable lateral modes is important to composite applications. We have reported that the lateral modes and mode coupling can be strongly affected by the choice of constituent materials, and the specific structure designed.<sup>[1]</sup> The method using interferometry to investigate the thickness and lateral vibration modes was also reported.<sup>[2]</sup>

In this presentation, the properties of 2-2 and 1-3 composites of different volume fractions with random pitches and constant kerf, and the 2-2 composites in which both pitches and kerfs are random have been compared with regular composites. It has been discovered that random dicing structure consisting of random pitches with either constant or random kerfs, will be able to significantly suppress the lateral resonances.

The effects of undesirable lateral modes on the performance of transducers, and how these effects can be reduced or suppressed by composite structure and constituent material have been also investigated. It has been shown that transducer performance is well correlated with the dynamic behavior of composite. A random diced structure will be able to suppress the lateral modes in composite, hence, the transducer performance will be improved.

The financial support by the ONR through the Grant No. N00014-91-C-0229 is greatly appreciated.

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[1] Yuan et. al., "Experimental Studies of Piezo-composites, Part I: Comparison With Theoretical Models." ONR Transducer Materials and Transducers Workshop. State College, PA. 1995.

[2] Yuan et. al., "Experimental Studies of Piezo-composites, Part II: Dynamic Behavior." ONR Transducer Materials and Transducers Workshop. State College, PA. 1995.

# RESONANT MODES, ACOUSTIC IMPEDANCE, AND SURFACE VIBRATION PROFILES OF PERIODIC PIEZOCERAMIC-POLYMER COMPOSITE PLATES

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As a transducer material piezoceramic polymer composites offer many advantages over the conventional single phase ceramic and polymeric materials. Being a diphasic material, the properties of a composite can be tailored over a wide range and due to the coupling between the two constituents, there are many new features which do not exist in any single phase material. In this talk, the results of a combined analytical and experimental investigation of many properties such as the resonant modes, the acoustic impedance, and the surface vibration profile of a period piezocomposite plate under different boundary conditions (both in air and in fluid) are presented. These properties are important for the design of ultrasonic composite transducers. The new approach used in the analytical model allows us to quantitatively predict how various modes and their coupling to the thickness resonance change as the aspect ratio of ceramic plate changes. The results also show that the input acoustic impedance between a fluid or solid medium and a composite is not a constant but decreases with frequency at frequencies below the stop band edge resonance. In addition, the value of the input acoustic impedance for a composite plate depends on the medium it is in contact. Those findings have important implications to the design of the composite, matching layer and backing for a composite ultrasonic transducer.

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# Finite element study of varied impedance matching layer properties

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## ABSTRACT

In order to increase the transduction of ultrasonic signal from a high impedance piezoelectric transducer to a low impedance medium, an intermediate matching layer is used. The introduction of this matching layer will cause a shifting of resonance frequency and may introduce additional surface modes. The transducer designer would then like to fully understand the effects from varying the matching layer properties (i.e. thickness and acoustic impedance).

For this reason we have studied the effect of increasing and decreasing thickness of the quarter wavelength matching layer and changing its acoustic impedance on a 1-3 PZT-5H composite transducer using finite element analysis. The results are also compared to a Krimholtz, Leedom & Matthaei (KLM) model. The study is focused on the resonance frequency and the displacement profile on the surface of the matching layer.

1996 ONR Materials and Transducers Workshop

**PIEZOELECTRIC POLYMER APPLICATION IN SOLID-STATE INTRACORONARY  
ULTRASOUND (ICUS) HEART CATHETER:**

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A successful medical application, which has been cleared for marketing by the FDA, is described using a piezoelectric polymer in a solid-state intracoronary ultrasound solid state (ICUS) heart catheter. EndoSonics Corporation produces an imaging system with various heart catheters, which incorporate an ICUS transducer into the catheter tip. The 1.2 mm OD transducer design consists of a ceramic base, 4 integrated circuits, a PVDF-TrFE copolymer, a biocompatible/acoustic matching encapsulation, various metallization layers, and 7-wire microcable. Dynamic aperture array acoustic technology is employed. The 20+ MHz transducer and system processes 1500 acoustic vectors to produce a 2D digital reconstruction. Three dimensional capabilities are also possible with the digital format. The system processes signals at 500 million operations per second. ICUS provides a an image perpendicular to the catheter axis and has a slice thickness of less than 1 mm. The artery view shows a 360 ° tomographic field with an axial resolution of 80  $\mu$ m.

Several examples of heart disease are shown with ICUS and angiography (an external x-ray technique) to interpret heart disease. Current improvements of ICUS include higher image resolution, color, user-friendly WINDOWS interface, and on-going development of combined diagnostic and therapeutic devices. Catheter-based therapeutic techniques, along with diagnostic ultrasound, offer cost-effective solutions to address heart disease in an ever-changing health care environment. Minimally invasive, catheter-based diagnostic and therapeutic procedures have significantly reduced the need for open heart surgery for many patients. Ultrasound imaging has applications in other anatomical areas such as peripheral vessels, urology, gynecology, and neurology.

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***25 March 1996***

***Afternoon  
Presentations***

1996 ONR TRANSDUCER MATERIALS AND TRANSDUCERS WORKSHOP

ACTIVE CONTROL OF STRUCTURALLY RADIATED SOUND USING AN  
INTEGRATED PIEZOELECTRIC DOUBLE AMPLIFIER SKIN

by

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This paper will describe the development and use of a double amplifier, piezoelectric based skin for the active reduction of sound radiation from vibrating structures. For large active reduction of sound at low frequencies in a light acoustic medium such as air, the active source must "pump" as much fluid as possible. Thus the use of piezoelectric devices in this application essentially involves amplifying the device normal displacement(at the cost of force which is not critical for air). The presentation will first detail the configuration of the skin and numerical analysis of its acoustic behavior. The numerical results indicate that even though the surface motion is complex, at low frequencies the active skin essentially behaves as a monopole source. Results of experiments will be presented which show broadband active sound control of radiation from a simple structural piston radiator of the order of 10-15dB when using an LMS control approach in conjunction with a far-field error microphone. Experiments on the more complex situation of panel radiated broadband sound as well as the integration of the pressure error sensors into the skin will then be considered.[Work supported by ONR]

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# DEVELOPMENT OF TRANSVERSELY ALIGNED PIEZOELECTRIC FIBER COMPOSITES FOR ACTIVE STRUCTURAL ACOUSTIC CONTROL

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Piezoelectric fiber composites have been developed as an anisotropic actuator to be integrated with composite host structures for a variety of applications. The composite form of the actuator offers advantages in strength, reliability, and conformability, as well as orthotropic actuation in comparison with traditional monolithic ceramic actuators. More recently, the interdigitated electrode pattern has been introduced, greatly enhancing the performance of the actuators. Other developments include electromechanical models and demonstration of induced shear capabilities in manufactured active laminates. Research efforts have continued to improve the materials, the manufacturing, and the characterization of the interdigitated electrode piezoelectric fiber composites (IDEPFC).

New hybrid matrix studies have investigated the use of carbon black as a filler which may be used in conjunction with PZT and other additives to improve the dielectric. Frequency dependent effects and impedance matching are also being investigated. In the area of manufacturing, reinforcing glass filaments have been integrated within the actuator in order to improve the strength and toughness of the composite. The continuous, 9  $\mu\text{m}$  diameter, S-glass filaments surround the larger and more brittle ceramic fibers, providing an efficient load transfer mechanism around discontinuities. A complete set of characterization tests was developed to evaluate the performance of the baseline IDEPFC actuators, glass-reinforced actuators, and actuators embedded in fiberglass laminates. The tests are designed to evaluate the electromechanical properties of the actuators and to monitor the performance of the actuators in application-driven experiments. These include long-term electrical and mechanical fatigue testing and actuation under static mechanical loading.

The final section of this work presents a preliminary design for an active composite plate for reducing broadband sound radiation and transmission using feedback and adaptive feedforward control. The composite plate incorporates several active, anisotropic plies which are segmented using patterned interlaminar electrodes. This arrangement allows for the independent control of structural modes. A model which accounts for the structural and acoustic modes has been implemented and used to optimize the location of the active subplies to minimize the global sound radiated from the structure. Open loop predictions are given for the acoustic actuation authority of the active panel. Future work will include the manufacture and testing of the plate, as well as the application of modern control methodologies.

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## MONOLITHIC ACCELEROMETER FOR SMART ACTUATOR APPLICATIONS

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This paper describes a fabrication approach for high-sensitivity low-cost accelerometers. This approach offers the potential for producing inexpensive highly-sensitive accelerometer arrays (for example, for underwater acoustic velocity sensing applications), or intrinsically combining accelerometers as a dense array within an actuator (for active acoustic control and smart materials applications).

Conventional piezoelectric accelerometers consist of a mass supported by a piezoelectric element with electrodes. While simple in concept, the fabrication of a high quality measurement accelerometer can be labor intensive. The monolithic accelerometer approach described in this paper fabricates the mass and piezoelectric element from one single piece of piezoelectric ceramic. This offers a number of advantages over the conventional approach. The fabrication is simpler since separate construction of the mass is eliminated, as is the process of bonding the mass to the sensor. Reliability is improved since the bond uncertainty is removed. Thermal shock noise associated with the bond is also eliminated. Additionally this accelerometer design is suitable for inexpensive large-quantity fabrication using an injection molding process such as that of Material Systems Inc. (MSI).

Several such monolithic accelerometers have been designed at NRL and fabricated by MSI. Laboratory measurements have found their performance to be highly predictable and competitive with good instrumentation accelerometers. For example, a typical accelerometer for NRL smart actuator studies is 6.7 mm in radius and 7 mm tall. It has a flat sensitivity (150 mV/g) and a low noise floor (200 ng/ $\sqrt{\text{Hz}}$ ) over the measured frequency band 500 Hz to 20 kHz. The high frequency limit is higher than 30 kHz, and the measured transverse sensitivity is only 3% of axial.

This approach is particularly attractive for fabricating an internal accelerometer array within an actuator. MSI currently produces an injection molded 1-3 composite actuator. In the same processing step, MSI can injection mold an accelerometer array, such that sensing and actuation functions are intrinsically combined into one co-formed inexpensive transducer array.

Combined sensor - actuator prototypes have been constructed by NRL and MSI, and their characteristics evaluated. Issues encountered include particularly mechanical and electrical crosstalk and system resonances. Results are presented which show that the combined transducer has predictable properties and is well suited for use in active-control applications.

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## A CONCURRENTLY ENGINEERED ADAPTIVE COMPOSITE PANEL

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The objective of this research was to design, fabricate, and test an adaptive, quiet, composite sandwich panel capable of producing substantial sound power reductions over a frequency band ranging from 0 to 750 Hz. A concurrent engineering development strategy that integrates materials, design, analysis, and fabrication, controls, and testing disciplines was used to develop the adaptive panel. The composite panel consists of inner and outer composite laminate skins separated by an array of cascaded flextensional transducers embedded in a structure grade core material. Each of the cascaded flextensional transducers consists of an inner and outer flextensional frame where the outer frame is driven along its major axis by two co-fired, multi-layered piezoceramics stacks. The extensional motion of the piezoceramic stacks are amplified and converted into flexural motion of the frames. The resulting amplified motion is used to drive the surface of the composite skins. The induced motion normal to the surface of the composite skins was used to control the panel's radiated sound power. Finite element analyses were performed to predict the performance of both the cascaded flextensional transducers and the composite panel. Excellent correlation was obtained between the experimental measurements and the predicted response. Measurements were made in a reverberation chamber with the panel active and inactive to determine the levels of sound power reduction achievable with the adaptive, composite sandwich panel. Sound pressure level reductions up to 25 dB over a frequency band ranging from 0 to 750 Hz were achieved by driving the piezoceramic stacks at voltages ranging from 55 to 100 volts rms.

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**25 March 1996**

***Afternoon  
Poster Presentations***

# Processing Control During the Fabrication of PMN Powders by a Liquid-mix Process

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## Abstract

A liquid-mix process was developed to synthesize perovskite  $Pb(Mg_{1/3}Nb_{2/3})O_3$  (PMN) and  $0.9Pb(Mg_{1/3}Nb_{2/3})O_3 - 0.1PbTiO_3$  (0.9PMN-0.1PT) powders. The processing parameters to prepare the PMN precursor with uniform mixing of the cations at the atomic scale were studied in detail. Phase development studies showed that the pyrochlore phase began to crystallize at  $500^\circ C$  with the perovskite phase appearing at  $\approx 600^\circ C$ . At intermediate temperatures the precursor yields a very fine mixture of pyrochlore and perovskite phases. The transformation of pyrochlore to perovskite was complete at  $1000^\circ C$  for pure PMN powders, but the pyrochlore phase remained at  $1000^\circ C$  for 0.9PMN-0.1PT powders, probably due to a higher level of lead loss. Nevertheless, both powders calcined at  $600^\circ C$  were sintered to nearly full density at  $1200^\circ C$  with the pyrochlore phase completely transforming into perovskite phase in situ. A bimodal densification behavior was observed in the sintering kinetics for both powders, which demonstrated the impact of the phase transformation on the sintering behavior. This strategy of sintering a fine mixture of pyrochlore and perovskite powders and forming the desired perovskite phase in situ was proven to be useful and promising.

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## A COMPARATIVE STUDY ON THE KINETICS OF HYDROTHERMALLY DERIVED PEROVSKITE-TYPE MATERIALS

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A study was conducted to determine the minimum conditions (eg. amount of pH adjusting agent and temperature) to hydrothermally precipitate perovskite-type materials that could be synthesized from acetate metal-cation feedstocks and nano-sized oxides or submicron amorphous hydrous oxides. The conditions (eg. temperature, reagent, input concentrations, and the amount of pH-adjusting agent) for hydrothermal precipitation of alkaline earth (i.e. Ba and Sr) titanates and lead titanate-zirconates were modeled in their respective thermodynamically stable phase pure regions. A comparison between the theoretically modeled and experimental conditions were made. The model predicted a reaction at room temperature for all the compositions. In addition, the amount of pH-adjusting agents (usually KOH) was determined from the amount of input concentration of starting materials. Pure phase PT, PZ and PZT were experimentally found to precipitate at temperatures between 100 and 200°C. However, barium and strontium titanate were experimentally found to precipitate at lower temperatures (between 40 and 200°C). Furthermore, the experimental conditions for pyrochlore lead titanate were found experimentally to exist below the conditions for pure lead titanate. It can be concluded that the thermodynamic model can be used to predict the concentration of mineralizer but the reaction temperature is dominated by kinetics.

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## NOVEL CHEMICAL PROCESSES FOR MANUFACTURING FINE-SCALE PIEZOELECTRIC MATERIALS

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In September 1995, an ONR supported Phase I STTR program was initiated as a collaboration between NexTech Materials, Ltd., a small advanced ceramic manufacturing firm, and Pacific Northwest National Laboratory, a Department of Energy research organization. The goal of the program is to develop economical and innovative chemical processing technology for manufacture of fine-scale piezoelectric materials. If successful, our approach will enable NexTech Materials to build a small to medium scale production facility with a relatively small capital expenditure. The production cost will be competitive with conventional and alternative chemical process technologies used for making high quality ceramic powders. Furthermore, the products made using the new process will be clearly differentiable from competitive products in regard to electrical and mechanical performance, and reliability in very fine-scale, high frequency-driven structures.

The product selected for investigation is a lead-lanthanum zirconate-titanate (PLZT) composition with high dielectric constant ( $K_{33}^T = 3500$ ) and electromechanical coupling ( $Q = 47$ ). Applications for the material include transducers for high resolution ultrasound systems and high performance actuators. Two processes are being investigated: batch hydrothermal synthesis, and Rapid Thermal Decomposition of Solutions (RTDS). Both processes involve relatively low temperatures, less than 350°C, and elevated pressures. The hydrothermal process has been previously described by the presentor. In the present work, the raw materials employed are lead oxide, lanthanum nitrate, zirconium oxynitrate, and titanyl nitrate prepared by a low cost route. The RTDS process entails the very rapid (less than 1 minute) decomposition of a mixed nitrate salt solution. The major advantage of the RTDS process over batch hydrothermal synthesis is a significant reduction in capital cost.

The initial results of the study have allowed for a comparison of physical and chemical characteristics of the ceramic powders, and the dielectric and piezoelectric properties of derivative fine-scale ceramic materials. A comparison of processes and properties is given in the table below. Electrical properties will also be presented.

COMPARISON OF MATERIALS BY POWDER METHOD

Method:	Conventional	Hydrothermal	RTDS
Reaction Conditions	900 C	300 C, minutes	350 C, seconds
Avg. Particle Size, $\mu\text{m}$	1-2	0.1-1.0	<0.5
Surface Area, $\text{m}^2/\text{g}$	<4	4-20	>20
Typ. Crystallite Size, nm	1000	100-200	<20
Powder XRD	PLZT solid sol'n	PLZT solid sol'n	amorphous
Sintering Temperature, C	1250-1300 C	1100-1200 C	in progress
Sintered Density, g/cm <sup>3</sup>	7.69	7.79	in progress
Avg. Grain Size, $\mu\text{m}$	3-10	1-3	in progress

## **Processing and Structure-Property Relationships for Fine Grained PZT Ceramics**

**T. R. SHROUT, C. A. RANDALL, N. KIM, W. CAO, and W. S. HACKENBERGER**  
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This study investigates the structure-property and processing relationships for submicron PZT ceramics fabricated using both conventional and pressure assisted sintering. PZT ceramics with average grain sizes less than 0.5  $\mu\text{m}$  were produced using a combination of high energy milling, B-site precursor calcination, and liquid phase sintering via excess PbO additions. Further reductions in grain size to 0.1  $\mu\text{m}$  were achieved with hot pressing and hot isostatic pressing. Piezoelectric and dielectric activity are known to decrease with decreasing grain size due to clamping effects resulting from the increased density of grain boundaries. Transmission electron microscopy (TEM) revealed a departure from the accepted parabolic scaling law relating grain and domain size. Furthermore, fewer domain variants were found in submicron grains compared to what is typically observed in coarser grained material ( $\geq 1 \mu\text{m}$ ). The reduced variants would limit the amount of remnant polarization and the associated piezoelectric coefficients. TEM also revealed that transgranular domain coupling was dependent on the elastic stress state at grain boundaries.

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## SINTER-FORGING OF FINE GRAINED DIELECTRICS

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Densification rates of ceramics have been shown to increase without significantly increasing grain growth by applying a shearing force during sintering. Sinter-forging is a dieless process that applies a uniaxial force to a sample while it is sintering. Through a complex process that includes particle rearrangement, the larger interagglomerate pores of the system are eliminated. Hence, the pore size distribution can be made smaller and tighter before the ceramic sinters to full density.

Our presentation centers on the results obtained in a prototypical system of industrially produced hydrothermal precipitated BaTiO<sub>3</sub>. Sinter-forging was carried out by applying uniaxial pressures of 100 to 400 MPa to die pressed samples in a temperature range of 800 to 1100 °C. Submicron grain sized BaTiO<sub>3</sub> samples with densities above 90% have been produced. Pressureless sintering of this BaTiO<sub>3</sub> powder produced a noticeable amount of abnormal grain growth when sintering to high densities. Sinter-forging reduced or eliminated abnormal grain growth by sintering at lower temperatures for shorter times.

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## LOW TEMPERATURE PROCESSING OF LEAD ZIRCONATE TITANATE PIEZOELECTRIC GLASS-CERAMICS

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Since 1991 we have conducted research at the Colorado Center for Advanced Ceramics to determine the potential of developing ferroelectric glass-ceramic compositions which densify at low temperatures in glass powder form, crystallize ferroelectric phases, and can be electrically poled to produce useful piezoelectric properties. These characteristics have been developed in glass-ceramics consisting of lead zirconate-titanate crystallites in a lead silicate matrix. These PZT glass-ceramic compositions densify at low temperatures (<850°C) by viscous phase sintering of the starting glass powder at ≈450-550°C combined with liquid phase sintering at ≈750°C. This presentation will discuss optimization of the electrical properties by addition of SrO, Nb<sub>2</sub>O<sub>5</sub>, MnO<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub> to the glass compositions. The effect of Zr/Ti ratio varied across the morphotropic phase boundary will also be presented. Additions of SrO, Nb<sub>2</sub>O<sub>5</sub>, and MnO<sub>2</sub> were used to lower the Curie temperature, improve the ease of domain wall motion for electrical poling, and to increase the resistivity. Addition of Al<sub>2</sub>O<sub>3</sub> improved the glass formability. Varying the Zr/Ti ratio allowed the piezoelectric properties to be optimized with values of the piezoelectric charge coefficient  $d_{33}$  of 44 pC/N and piezoelectric voltage coefficient  $g_{33}$  of 33x10<sup>-3</sup> Vm/N. Details of the processing methods and resulting dielectric, ferroelectric, and piezoelectric properties will be discussed.

This research was supported by the Office of Naval Research and National Science Foundation.

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## INTELLIGENT PROCESS MODELS FOR BETTER SYNTHESIS PROCESSES FOR FERROELECTRICS

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Activities at Rutgers are focused on developing intelligent synthesis models that lead to the successful preparation of phase pure homogeneous perovskite materials (e.g., BST, PZT, PMN) utilizing conventional as well as advanced methods such as hydrothermal synthesis. The ultimate objective is to define engineering principles that facilitate time effective intelligently designed experiments rather than time-consuming empirical experimentation. Utilizing advanced thermodynamic modeling, we are able to identify the concentration, temperature and pH at which phase pure materials can be hydrothermally prepared. Thermodynamic modeling is also a powerful tool for other synthesis methods such as coprecipitation, where metal hydrous oxides must be precipitated with controlled stoichiometry. Furthermore, minimal leaching of ceramic powders in aqueous media can be engineered for processes such as powder washing, milling or tape casting. Another recent advance is the development of the Concentric Shell Model of Mixedness (CSMM), which can be employed for optimizing the solid state reactivity of a particulate mixture. The model can determine how the mixedness of a particulate mixture of a specific composition is influenced by the powder characteristics such as particle size, particle size distribution, molar volume and packing density of the powder bed. Criteria can be employed to design experiments through mixedness engineering selecting precursor powders, packing densities, and methods for mixing, milling, and size separation that will yield phase pure, chemically uniform solid state reaction products.

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# Synthesis of Modified PbTiO<sub>3</sub> Powders and Fibers by a Liquid-mix Process

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## Abstract

Modified PbTiO<sub>3</sub> powders and fibers were fabricated using a liquid-mix process. The processing parameters, such as calcination temperature, ball milling conditions, and sintering temperature, were correlated to the degree of powder aggregation, phase development, density, grain size, and microstructural uniformity. Results indicated that the degree of aggregation was strongly dependent on the calcination temperature, and less dependent on the milling solvent and pH values. Powders with an extremely small particle size ( $\approx 0.1\text{-}0.2\mu\text{m}$ ) were obtained by calcining the precursor resin at 450°C, while achieving the desired perovskite structure. Powder compacts made from this powder were sintered to nearly full density at 1200°C with a uniform microstructure. Through proper control over the rheological behavior of the precursor with the addition of a small amount of polyacrylic acid, continuous precursor fibers were drawn and oxide fibers were obtained after a two-step heat treatment at 600°C in N<sub>2</sub> and air. Both hollow and solid fibers were observed. The strength of the fibers were high enough to be handled, yet, not high enough to enable the fabrication of 1-3 composite structure. Further improvement on the processing control needs to be done to perfect the quality of the fibers. Nevertheless, the feasibility of the liquid-mix process for fabricating modified PbTiO<sub>3</sub> fibers was well demonstrated.

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1996 ONR TRANSDUCER MATERIALS AND TRANSDUCERS WORKSHOP  
ABSTRACT

TAPE CASTING OF POWDER MATERIALS  
USING INTELLIGENT PROCESS CONTROL METHODS

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Tape casting is a complex process with highly coupled process variables and materials parameters and is ideally suited to the implementation of intelligent processing of materials (IPM) principles. Current practice uses limited sensors for tape transfer speed control and drying unit temperature control in spite of large variations in process parameters, and significant coupling between casting and drying operations.

A tape casting machine (4 meters long) has been designed and constructed for a series of experiments on real-time, intelligent control. The machine includes an actively controlled casting unit, drying unit, and tape transfer unit, as well as instrumentation for advanced process monitoring and intelligent control. The initial controller developed combines adaptive control with a rule-based expert system and graphical user interface suitable for testing of a variety of materials and the creation of an offline materials data base.

Results to-date include:

- Successful casts of a variety of materials including AlN, SiC and PZT. Slurries based on solvents including toluene, alcohol and water were used; dried tape thicknesses ranged from 25-800 $\mu$ m; controlled drying to improve the quality of thick tapes was demonstrated.
- Real-time intelligent control has been demonstrated using both model-based and closed-loop control of the green (as-cast) tape thickness. The model used for the casting process<sup>1</sup> is based on Newtonian fluid flow and predicts tape thickness as a function of several material and machine parameters. Results show that improvements in the setup time, consistency, and flexibility of the process are attainable. A recently published model<sup>2</sup> which accounts for non-Newtonian fluid flow is currently being examined and tested to determine its applicability to conditions such as thick tapes, high solids loading, rapid drying.
- Thin PZT tapes have been utilized to produce shaped transducers using a "cut-and-stack", multilayered manufacturing technique.
- Preliminary studies have shown the effect of doctor blade profile on fluid flow. Modeling showed that blade profile had a significant effect on turbulence of the slurry as it exits the reservoir. Experiments showed the effect of blade profile on green tape thickness.

Research is ongoing to implement computer integration for intelligent process control, and to develop real-time control methods which incorporate advanced materials processing knowledge to improve the quality and flexibility of the tape casting process. Results suggest that machines utilizing computer integration and intelligent process control can offer a substantial improvement over standard industrial tape casting machines. In addition, such machines may offer the ability to cast materials and/or geometries not possible with standard machines.

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## A NOVEL APPROACH TO FABRICATING HIGH $Q_m$ PZT

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It is well-established that PZT can be hardened or softened by adding dopant elements. The use of a dopant phase is perhaps less common. In this study, the effect of 5 at% of the dopant pyrochlore phase  $Sr_2Sb_2O_7$  was investigated. The effect of Mn additions (0.1, 0.3, and 0.5 wt%) in the form of  $MnO_2$  was also investigated. It was postulated that the pyrochlore phase might pin the domains within the perovskite grains, thus increasing  $Q_m$ . It was further postulated that compositions with both the pyrochlore phase and the Mn additions should yield even greater  $Q_m$  values. Properties of interest,  $k_p$ ,  $Q_m$ ,  $\epsilon$ , and loss, were measured by the resonance method. The overall goal was to determine an improved composition for a surface acoustic wave device application.

# STUDIES ON THE ELECTROMECHANICAL PROPERTIES OF PbTiO<sub>3</sub>-BASED CERAMICS

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Lead titanate (PbTiO<sub>3</sub>) exhibits many interesting characteristics including highly anisotropic piezoelectric properties, a high piezoelectric voltage coefficient due to its lower dielectric constant, a high Curie temperature, a high spontaneous polarization, a low aging rate, and a relatively low mechanical quality factor. These attributes make PbTiO<sub>3</sub>-based materials attractive for many bulk, thin film, composite, and single crystal applications.

In our lab we have been performing several studies on modified lead titanate compositions with the purpose of determining whether simultaneous Ca and Sm additions result in desirable piezoelectric and dielectric properties similar or superior to previously-prepared systems. Results showed that the (Pb<sub>0.88-x</sub>Ca<sub>x</sub>Sm<sub>0.08</sub>)(Ti<sub>0.98</sub>Mn<sub>0.02</sub>)O<sub>3</sub>; x = 0.11, 0.13, 0.15 and 0.17 system exhibits normal ferroelectric behavior, with most properties similar to other modified lead titanates. The unique attribute of this system appeared for the x=0.13 and 0.15 compositions, which exhibited k<sub>t</sub>'s  $\geq$ 57%.

In another series of studies the phase diagram of the (Pb<sub>0.85</sub>Sm<sub>0.10</sub>)(Ti<sub>0.98</sub>Mn<sub>0.02</sub>)O<sub>3</sub> - BiFeO<sub>3</sub> system was studied by measuring the dielectric behavior as a function of temperature, coupled with room temperature XRD and measurements of d<sub>33</sub>, k<sub>t</sub>, and k<sub>p</sub>. A solid solution forms over a wide range between the modified PT and the BF, with a mixed phase region occurring between 57.5 and 60%. The maximum K', k<sub>t</sub>, k<sub>p</sub> and d<sub>33</sub> were exhibited in this MPB region. Some of the studied compositions exhibit electromechanical properties suitable for transducer applications, over a compositional range where the K' varies widely.

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***26 March 1996***

***Morning  
Presentations***

1996 ONR TRANSDUCER MATERIALS AND TRANSDUCERS WORKSHOP  
ABSTRACT GUIDE

**TITLE: A 128x4 CHANNELS 1.5D CURVED LINEAR ARRAY FOR MEDICAL IMAGING**

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1.5D transducer arrays represent a promising improvement in ultrasonic B-mode imaging. The reduction of the slice thickness and the correction of phase aberration are potential applications of 1.5D arrays, while keeping the number of electronic channels on ultrasound beamformers between 64 and 256.

We present the design and manufacturing, using a high density interconnection process, of a 1.5D curved linear array with 128x4 independant elements, allowing dynamic focusing in the elevation plane.

We have compared the performances of different elevation samplings versus two factors of merit. Little differences between Fresnel like sampling and equiimpedance type sampling have been observed. Consequently, the sampling choice has been dictated by the compatibility with the system transmitter and receiver.

We have then manufactured a 128x4 channels curved linear array for medical imaging, using a novel interconnection technology. This collective technology is well suited to connect a large number of elements along the elevation direction and is shown to be compatible with large bandwidths needed to obtain high quality imaging.

The experimental results conducted in water tank are showing improved elevational resolution on a wider range than 1D probes.

Further experiments on phantoms using a 64x4 beamformer and clinical images are planned in order to compare this prototype with a reference probe having same azimuthal pitch, radius of curvature, center frequency and bandwidth but constant elevational aperture and will qualify the potential of such 1.5D probes to ease the diagnosis of physicians.

## PHASE ABERRATION CORRECTION IN TWO DIMENSIONS USING A DEFORMABLE ARRAY TRANSDUCER

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Phase aberrations due to tissue inhomogeneities degrade medical ultrasound images by disrupting the ultrasound beam focus. Currently, several phase correction algorithms are implemented by adjusting the electronic phase delays used to steer and focus the ultrasound beam. This means that a two-dimensional (2-D) array is necessary to completely correct 2-D aberrations in tissue. However, 2-D arrays are a complex option due to their large number of elements and poor sensitivity. Instead of using a full 2-D array, a new technique is proposed which uses actuators to fabricate a deformable transducer of significantly fewer channels for 2-D phase correction. Phase correction in azimuth is achieved by altering the electronic phase delay of the element. However, phase correction in elevation is achieved by tilting the element in elevation with a low frequency piezoelectric actuator. Comparison of simulations of the new phase correction transducer versus the conventional phase correction technique have shown that a deformable  $1 \times N$  or  $2 \times N$  transducer can approach the image quality of a  $4 \times N$  2-D array or greater. A prototype  $1 \times 32$ , 2.5 MHz, array with eight low frequency piezoelectric actuators has been constructed such that every four ultrasonic transducer elements in azimuth are mounted on one independently controlled actuator. This prototype transducer was used to test the ability of the deformable array to produce real time phased array scans and to simulate on-line phase correction by tilting the elements in the elevation direction with the actuators. A  $2 \times 32$ , 3.5 MHz, deformable array was also constructed and tested. This array relied on RAINBOW actuators for both the physical displacement and generation of ultrasound.

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1996 ONR TRANSDUCER MATERIALS AND TRANSDUCERS WORKSHOP

DYNAMIC IMAGING OF PIEZOCOMPOSITE TRANSDUCERS

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Advanced piezocomposite transducers are continuously being designed and constructed for a multitude of applications. Most of their characterization involves the electrical resonance spectra response or their far field behavior. In both cases it is the average effect of the surface displacement that is measured. With Laser Heterodyne Interferometry the direct surface displacements of ultrasonic transducers can be measured. In this work we will present surface displacement measurements on 2-2, 1-3 periodic and random structures. These transducers have been fabricated by Echo Ultrasound Inc. and by Materials Systems Inc. Finally, in collaboration with Penn. State Univ., a direct comparison between actual surface displacements measurements obtained using laser interferometry and finite element modeling will be presented.

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***Morning  
Poster Presentations***

## FINITE ELEMENT MODELING OF A 2-DIMENSIONAL TRANSDUCER ARRAY

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A 10,000 element, 3 MHz transducer array, backed with integrated transmitter and receiver electronics, was proposed by E. Belcher of the Applied Physics Laboratory, University of Washington, as the Audio-Visual Converter (AVC) in a acoustic lens focused, diver-held sonar system. A feasibility study was conducted in three-dimensions to determine relevant performance parameters of candidate piezoelectric composite array structures, including electrical impedance, pressure transfer function, cross-talk, displacement profile, and beam profile. The array consists of 10,000, 1.0 mm diameter elements spaced 1.75 mm apart in a hexagonal arrangement. The study was conducted using PZFlex, a time domain based finite element code from Weidlinger Associates. The three-dimensional scale of the problem, examining crosstalk levels out to the third nearest neighbor, required a larger model size than heretofore attempted in ultrasonic transducer arrays.

A trade study was initially conducted to select the most promising of several candidate structures based on injection molding technology available from Materials Systems, Inc., Littleton, MA. Elements consisting of seven rod clusters in a soft polymer matrix were analyzed, and preferred matching layer materials, shapes, and thicknesses were determined. The preferred configuration utilized cylindrical silver-epoxy matching layers a quarter wave thick. Other configurations, including filled epoxy and aluminum matching layers, continuous or cylindrical, and 3/4 and 5/4 wavelength thick, were analyzed in order to achieve the best beam profile. Excellent beam shape and cross-talk performance better than -40 dB were obtained in the preferred configuration and will be described in the presentation.

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## NONLINEAR MODELING ISSUES FOR ULTRASOUND TRANSDUCERS

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A number of nonlinear phenomena can occur in ultrasound applications involving the transducer (e.g., high piezoelectric driving voltage and electrostrictors), the propagation path (e.g., shocking and cavitation), or both. Algorithmic or phenomenological difficulties can make modeling this behavior tricky. For example, algorithms relying on superposition, e.g., a frequency domain approach, are of limited use, and shocking phenomena produce high gradients or harmonics that are difficult to resolve numerically.

An algorithmic approach that works well in practice for a variety of nonlinear phenomena is the incrementally linear approximation applied to a time-marching scheme that integrates the space-discretized differential equations. At each time step the mechanical, thermal or electrical properties of each medium are updated based on instantaneous or cumulative effects due to pressure, strain rate, temperature, electric field, etc.

This talk reviews examples of nonlinear modeling found in therapeutic ultrasound applications. Simulations are done using the time-domain finite element code, PZFlex, which has been described in previous review meetings and is used commercially for medical ultrasound transducer design. Nonlinear illustrations include focused ultrasound and shock propagation in water, localized heating due to temperature-dependent absorption in propagation media, and progressive "cavitation" and scattering caused by elevated temperatures in the media.

Although we have not applied this approach to nonlinear transducer behavior, it will solve the associated phenomena equally well. Obvious applications include voltage or strain dependence of piezoelectric properties, which are only linear for small signals, and electrostrictors, which are inherently nonlinear (strain proportional to the square of polarization). The question is whether there is enough data and/or interest to pursue this class of nonlinear models.

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# Performance Analysis of a Low-Frequency Barrel-Stave Flextensional Projector

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A number of barrel-stave flextensional projectors for operation below 1 kHz in seawater have been built and tested at the Defence Research Establishment Atlantic (DREA) [1]. These high-power flexural-mode sources radiate sound efficiently at their resonance frequencies. Since they are small compared to the wavelength of the sound that they produce at resonance, these projectors are essentially omnidirectional. The purpose of this paper is to describe the construction of one of these projectors, to report the key measured acoustical performance parameters, to present an electroelastic finite-element model for the projector, and to illustrate the utility of finite-element techniques as a transducer design tool.

For the construction section, we include details on projector geometry and the materials used for the individual components. The performance parameters of interest are the transmitting voltage response, electrical admittance, mechanical quality factor, and electroacoustic efficiency.

The PZFlex [2] finite element analysis is performed in two stages. First, the piezoelectric ring stack is analyzed iteratively, starting with "textbook" values for piezoceramic material properties, and progressively modifying these properties to account for glue joint compliance and manufacturer's unique ceramic formulation. Second, the complete submerged projector is using an axisymmetric model. Calculated performance parameters are compared to measured ones. Finally, issues related to three-dimensional versus axisymmetric modeling for parametric studies are discussed.

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***Morning***  
***Poster Presentations***

## DEVELOPMENT OF ULTRA-FINE SCALE PIEZOELECTRIC FIBERS FOR USE IN HIGH FREQUENCY 1-3 TRANSDUCERS

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Research in obtaining high frequency transducers for ultrasonic imaging applications has prompted the need for fine scale composite structures. Fibers of La and Nb doped lead zirconate titanate have been fabricated for incorporation into 1-3 type composite transducers. Dense single fiber filaments with diameters from 10 to 60  $\mu\text{m}$  were produced using sol-gel technology. Gel fibers were extruded through a 100  $\mu\text{m}$  spinnerette and collected in a continuous fashion. Microstructural development during the gel to ceramic conversion was characterized under different sintering conditions. Submicron grains were found for fibers fired at temperatures lower than 1000°C with grains growing to 2-3 microns above 1200°C. The relationship between microstructure and dielectric properties has also been explored. The dielectric constant of PZT fibers with 1  $\mu\text{m}$  grain size averaged 700. Preliminary 1-3 composite processing and properties will also be presented. Funding provided by the Office of Naval Research Grant No. N00014-93-1054 and N00014-92-1391. The authors would also like to acknowledge the Whitaker Center for Medical Ultrasound.

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## FORMING AND SIZING OF FINE Pb(Zr,Ti)O<sub>3</sub> FIBERS

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We discuss the processing and sizing of Pb(Zr,Ti)O<sub>3</sub> fiber for use in transducer applications. Green Pb(Zr<sub>x</sub>Ti<sub>1-x</sub>)O<sub>3</sub> fibers, 80 to 100  $\mu\text{m}$  in diameter, were formed at Advanced Cerametrics, Inc. using the Viscous Suspension Spinning Process (VSSP). In this process, fine Pb(Zr,Ti)O<sub>3</sub> powder is intimately mixed with a polymer precursor. The powder + precursor mixture is spun through a spinneret into a coagulation bath to form fibers. The fibers are washed, dried, and collected on a spool. Yarns containing 10 to 500 individual fibers were collimated by applying a polymeric sizing to the yarns, then passing the yarns through sizing dies. Sized yarn tightness and flexibility was controlled by the sizing chemistry. To date, continuous sized yarns have been cut to short lengths, or woven in different architectures to yield novel microstructures. The short yarns were fired to produce straight rods of different diameters for "pick & place" piezoelectric ceramic/polymer composites. The woven structures were heat treated and backfilled with polymer to create composites with 1-3, 2-3, and 3-3 connectivity. After heat treatment, the diameter of the individual Pb(Zr,Ti)O<sub>3</sub> fibers was 10 to 20  $\mu\text{m}$ . Electromechanical characteristics of a number of composites were determined. Pb(Zr,Ti)O<sub>3</sub> VSSP fibers can be used to form fine-scale, large area piezoelectric fiber/polymer composites for use in hydrophones, transducers for medical ultrasonic imaging and non-destructive evaluation, and as sensors and actuators in vibration and noise control.

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## PZT MicroRod Composite Ultrasonic Transducers

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CeraNova is using it's MicroRod technology to develop an alternative to the slice and dice method of producing composite ultrasonic transducers. MicroRods, 120 $\mu$ m in diameter, are packed into a cylindrical form. Epoxy is infiltrated into the compact and cured. The composite is sliced perpendicular to the axis of the cylinder. Each slice becomes an individual transducer after electrodeing and poling.

This technique adds increased flexibility to the design of ultrasonic transducers. Transducers have been made using this method with PZT volume percentages between 50 and 80%. Packing can take geometries unattainable by slice and dice techniques such as hexagonal or random. Random packing of the MicroRods can reduce interpost resonances in these transducers. In addition, MicroRods of different sizes can be combined in a single composite. The size and shape of these composites can be varied greatly and is determined by a suitable choice of container for the MicroRod compact.

The support of the Office of Naval Research, contract number N0014-94-C0281, is greatly appreciated.

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1-3 PZT COMPOSITE TRANSDUCER RESEARCH AT ARL/PSU

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There has been considerable effort at ARL/PSU in using 1-3 PZT composite material in the design and fabrication of transducers and arrays. The primary material being used is the injection molding composite produced by Material Systems Inc. (MSI) consisting of 30% PZT-5H by volume, Shore D-80 hardness voided matrix material. Shaped sensors have demonstrated low sidelobe patterns and high source levels. The 1-3 composite panels have also been conformally shaped for a low cost high power source that can be easily fabricated. Design and test data will be presented for two large, shaped-aperture transmit arrays (50-100 kHz and 100-200 kHz) which were fabricated for NRL-SSC. These arrays produced 1 to 2 degree vertical beamwidths and horizontal beamwidths of 50 to 100 degrees at source levels greater than 200dBs. An acoustic dispersion lens was designed for the higher frequency array so that high power source levels and broad horizontal beamwidths could be achieved. In addition, a constant beamwidth transducer was designed and built, and some small units are being constructed and tested for use as wide bandwidth standard transmit/receive transducers to be used in calibration tests.

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## DETERMINING MATERIAL CONSTANTS, LOSSES, FREQUENCY DISPERSION, AND NON-LINEARITY WITH THE PIEZOELECTRIC RESONANCE ANALYSIS PROGRAM (PRAP)

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This poster/demonstration will introduce the Piezoelectric Resonance Analysis Program (PRAP) for Windows\*. PRAP is a Windows-base software package that allows for point and click determination of complex material constants governing the unloaded thickness, thickness shear, length, length thickness and radial electro-mechanical resonance modes. The program uses equations of the ANSI/IEEE standard<sup>1</sup> for determining material constants from piezoelectric resonances. It also determines losses for dielectric, piezoelectric, and elastic constants using curve fitting routines based on the work of Smits<sup>2,3</sup> and Sherrit et al.<sup>4,5</sup> The software has the provision for determining these complex material constants at the fundamental and higher order resonances. This allows for the determination of dispersion in each material constant over the range of resonances analyzed. The program may also be used to study the effect of a DC field on the material constants. Impedance spectra measured at different bias fields can be analyzed to determine the complex dielectric, elastic, and piezoelectric constants as a function of the DC field. PRAP also features routines to determine the circuit elements of Van Dyke's circuit model<sup>1</sup>, and the complex circuit models proposed by Sherrit et. al<sup>6</sup>. The demonstration will show examples of fits of the Impedance equations to simulated data for lossy, dispersive and non linear materials.

A copy of PRAP will be demonstrated and available for testing. Participants in the workshop are encouraged to bring their own Impedance spectra, including the geometry and density, which can be analyzed as part of the demonstration.

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<sup>1</sup>IEEE Standard on Piezoelectricity, ANSI/IEEE Std.176, 1987

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<sup>6</sup>S. Sherrit, H.D. Wiederick, B.K. Mukherjee, M. Sayer, To be published

\* Windows is a trademark of Microsoft Corporation

# Finite element study on random piezocomposite transducers

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## ABSTRACT

Piezocomposite materials have been widely used in low frequency transducers. In general the frequency range is less than 10MHz. The lateral pitch resonance caused by the periodic structure makes it unsuitable for very high frequency transducers. The key issue in making a high frequency composite transducer is how to suppress these lateral modes.

For this reason, we have studied the effect of randomness in a 2-2 composite. Two different modeling methods are used. One is the finite element method(ANSYS®), and the other is multisource T-matrix method. Results from both methods suggest that randomization can effectively suppress the pitch resonance. Generally speaking, 50-80% randomness is needed in order to destroy these lateral modes. The decoupling of the thickness resonance and lateral resonance also increases the electromechanical coupling coefficient for the thickness mode and shifts the thickness resonance frequency.

In order to understand the randomization effect on the acoustic field generated by the piezocomposite transducers, beam patterns have also been calculated by using ANSYS® and CHIEF.

## CLOSING THE LOOP IN TRANSDUCER MODELING<sup>†</sup>

### - Materials, Structures and Acoustic Field

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There are three stages in optimizing an acoustic transducer. First, one needs to fabricate good piezoelectric materials which can effectively convert electric energy into mechanical energy and vice versa. Second, one needs to engineer good composite structures that have good matching properties and can enhance the overall performance of the transducer material. The final step is to find the best configuration and geometry that can give the desired acoustic beam pattern. Through appropriate modeling processes, deeper understanding has been achieved for all the above three stages.

For the piezoelectric materials, the modeling effort has been on the formation of domain process since the domain processes give the dominant contribution (so called extrinsic contribution) to the piezoelectric and dielectric properties of piezoelectric ceramics. A 2-D model has been developed which could model the kinetics of the domain formation process in a ferroelectric system. Such a model can also be used to study the dynamics of domain switching process under an external electric field. For the second stage, finite element methods have been employed to study the transient behavior of a 1-3 composite. Through the finite element analysis, the vibration characteristics of the composite transducer become clear and the effects of size, ceramic volume ratio, ceramic aspect ratio and damping can be directly visualized. The FEM model can accurately predict the resonance frequency of composite transducers with complex geometry and provide all the modal structures, including the non-piezoelectric-active modes. In addition, FEM can precisely calculate the surface velocity map which could be fed into the Helmholtz integral to predict the acoustic beam pattern generated by the corresponding composite transducer.

Linking all three stages gives us a global modeling approach, i.e., **materials**→**structures**→**acoustic field**. It gives us a new dimension in transducer modeling. A clear follow-up work is to close the modeling loop by creating a feedback, i.e., **acoustic field**→**structures**→**materials**. Such a loop would provide a more comprehensive and powerful tool in the optimization of ultrasonic transducer designs.

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## AN ALTERNATIVE EQUIVALENT CIRCUIT FOR THE UNLOADED PIEZOELECTRIC RESONATOR

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An equivalent circuit model for the unloaded piezoelectric vibrator in the length, thickness, thickness shear and length thickness mode is presented. The model contains two branches, the motional branch and the static branch, like the lossless resonator model, but the capacitive and inductive elements are generalized by making each capacitance and inductance a complex constant. The losses (mechanical, dielectric, piezoelectric) associated with the vibrator are accounted for by the imaginary components of the circuit elements ( $C_0$ ,  $L_1$ ,  $C_1$ ). The model produced impedance curves that closely matched the impedance calculated by using equations derived from vibration theory and data from lead zirconate titanate samples and PVDF-TRFE copolymers. The calculation of the circuit parameters  $C_0$ ,  $L_1$ , and  $C_1$  from the complex elastic, dielectric and piezoelectric material constants is accomplished by the use of a set of complex frequency constants<sup>1,2</sup>  $f_s$ ,  $f_p$ , and the complex permittivity  $\epsilon$ . The model accurately represents the baseline dielectric behavior as well as the piezoelectric resonance around and below the fundamental resonance. Conversely, if the circuit parameters are known, the material constants can be derived by straightforward calculations without the loss of any information. In addition, the circuit values of the Van Dyke model may be calculated directly from the complex circuit parameters. It will be shown that this model fits both the impedance and admittance loci around and below resonance for a variety of materials and it displays reciprocity with the material constants for materials with very low Q. A similar model is presented for the radial mode.

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<sup>1</sup> S. Sherit, N. Gauthier, H.D. Wiederick, B.K. Mukherjee, *Ferroelectrics*, **119**, pp.17-32, 1991.

<sup>2</sup> S. Sherit, H.D. Wiederick, B.K. Mukherjee, *Ferroelectrics*, **134**, pp. 111-119, 1992.

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## EFFECT OF MECHANICAL STRESS ON THE ELECTROMECHANICAL PERFORMANCE OF PZT CERAMICS

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In many areas of application, piezoelectric actuator and sensor materials are often subjected to high mechanical stress fields and electric fields. Inevitably, the material properties will change with the external boundary conditions. In order to use these materials reliably and to optimize the performance of the material for a specific application, it is imperative to characterize and understand how the material properties change with these external conditions. In this talk, the results of a recent experimental investigation on the effects of mechanical stress on the electromechanical properties of several commercial available PZT piezoceramics, which are the most widely used piezoelectric actuator and sensor materials, will be presented. In this investigation, a new device was developed which allows us to characterize, for the first time, the piezoelectric coefficients of a material from both the direct and converse effects, all the matrix components of the elastic compliance, and the dielectric constant with mechanical stress under well defined mechanical and electric boundary conditions over a wide frequency range. As a result, the change of the electromechanical coupling factor of the material with stress can be determined. The results reveal that the transverse piezoelectric effect ( $d_{31}$ ) is very sensitive to the transverse stress and diminishes quite rapidly with the load in the transverse direction. In comparison, the  $d_{33}$  effect is much less sensitive to the load change. A comparison will also be made among the PZTs examined as to their actuator and sensor performance under different load conditions.

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ONR Transducer Materials and Transducer Workshop

Industrial Display Poster Abstract

"State of the Art Piezoelectric and Optical Components"

Russell S. Petrucci-Valpey-Fisher Corporation

Many of todays high frequency and high tolerance transducers require the best quality components. These high quality components help to minimize or eliminate performance variances at the "in process" and finished transducer stages.

Valpey-Fisher's Ultrasound and Optical Products has been supplying piezoceramics and single crystal materials for ultrasonic and optical applications for 65 years that meet and exceed most customer supplied specifications.

High density piezoceramics are supplied where grain size, elimination of voids and reliable dielectric tolerances are required. These materials have also been successfully fabricated as unbacked, unsupported elements up to 35 MHz. Piezoceramic types range from PZT, lead metaniobate, barium titanate and lead titanate.

Precision optics made of single crystal quartz, lithium niobate, sapphire, calcite and YAG have been routinely fabricated to attain critical flatnesses of 1/20 lambda with polished surfaces of less than .2 micron. These polishing techniques are available for use with most of the commercially available piezoceramic materials as well.

Valpey-Fisher has the capability to review industrial NDT, medical diagnostic and therapeutics, flow and level sensor, commercial, OEM and research applications to assist in selecting and fabricating the appropriate piezoelectric or optical material for these requirements. The vast knowledge and experience and techniques of fabrication employed by Valpey-Fisher are certain to enhance the finished products of any client requesting our assistance.

***26 March 1996***

***Afternoon  
Presentations***

## RECENT ADVANCES IN PIEZOCOMPOSITE MATERIALS, TRANSDUCERS, AND ARRAYS AT MSI

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A number of advances have been achieved in the fabrication of 1-3 piezocomposite materials and transducers. SonoPanel™ transducers with different PZT formulations, volume fractions, and polymeric matrices have been developed to meet the requirements of various underwater applications. Transducers have performed well under 6.9 MPa hydrostatic pressure and survived multiple explosive shocks. They can also be made to conform to nonplanar surfaces.

Several multiple element arrays have been fabricated from sheet 1-3 piezocomposite. Receive sensitivity equals or exceeds that achieved previously with 0-3 composite, with the added benefit of transmit capability. The feasibility of 1-3 cluster array elements as small as 1 mm diameter for 3 MHz acoustic imaging applications has also been demonstrated.

MSI is currently integrating both sensing and actuation into a single piezocomposite smart panel for active surface control. Multiple net-shape molded accelerometer elements have been designed and installed in 100 x 100 mm panels. Initial test results will be reported. Injection molded PZT bender arrays for air acoustic transducers are under development at MSI and in collaboration with MRL Penn State.

This work is supported by the Office of Naval Research and the Advanced Research Projects Agency.

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# 1996 ONR TRANSDUCER MATERIALS AND TRANSDUCERS WORKSHOP

## CONSTANT BEAMWIDTH 1-3 COMPOSITE TRANSDUCER

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Broad-beam, constant beamwidth transducers formed on a cylindrical arc can produce more power than conventional planar transducers with the same beamwidths. They provide more acoustic power because their area increases to produce broader beams instead of decreasing as with conventional planar transducers. These transducers also have the added benefit of having directivity patterns with constant beamwidth as a function of frequency. The utilization of 1-3 composite piezoelectric material enhances the performance and simplifies construction of constant beamwidth transducers due to its ability to be easily formed into a cylindrical arc. Test results show that a 1-3 composite constant beamwidth transducer provides almost 10 dB higher source level than a comparable 1-3 composite planar transducer. The constant beamwidth transducer also produces a 70° constant beamwidth directivity pattern from 75 to 300 kHz.

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# A SONAR APPLICATION OF 1-3 PIEZOCOMPOSITE MATERIAL

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Injection molded piezocomposite materials have significant potential for improving the performance of Navy and civilian acoustic systems. An array of 1-3 composite hydrophones has been manufactured and tested as a potential replacement for a currently used 0-3 composite (piezo-rubber) hydrophone developed for a high frequency sonar. Test results were obtained for the array panel alone and for the panel installed in a housing similar to the finished hydrophone module. The 4" x 8" panel was manufactured from injection molded PZT-4 piezocomposite and contains 40 elements.

Test results will be presented which include:

- Acoustic test results for the panel alone
- Effects of temperature and pressure on the acoustic performance of the panel
- Acoustic tests for the panel mounted in a housing

On the basis of these test results, a performance comparison with the currently used panel will be presented.

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## PROCESSING OF FINE-SCALE PZT FIBER AND FIBER/POLYMER COMPOSITES

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The processing of fine-scale PZT fiber and fiber/polymer composites for transducer applications is discussed. Emphasis is placed on inexpensive techniques that can form piezoelectric ceramic fiber and fiber/polymer composites with different connectivities and be easily utilized by composites manufacturers. Progress in several areas, including processing of PZT fibers, development of piezoelectric composites using PZT fibers and tapes, and formation of composites via a modified lost mold method and a solid freeform fabrication process, will be reported. The methods are summarized below:

(1) green PZT fibers, 10 to 30  $\mu\text{m}$  in diameter, were formed at Advanced Cerametrics, Inc. using the Viscous Suspension Spinning Process (VSSP). Yarns containing between 10 and 500 individual fibers were collimated by sizing the yarns with a polymer solution, and passing the sized yarns through a die. The binder system and die dimensions controlled the fiber dimensions and density in the yarn.

(2) The yarns were used to form PZT fiber/polymer composites with novel microstructures. Sized PZT yarns were bundled, fired, and backfilled with polymer to create composites with 1-3 connectivity. They were also woven into a cloth and a carpet structure.

(3) Stacked PZT tapes, as fine as 20- $\mu\text{m}$  thick, were backfilled with polymer to make composites with 2-2 connectivity. These structures yielded fine 1-3 piezoelectric ceramic/polymer composites when diced. Advantages of composites containing particulate fillers and composites with a PZT volume fraction gradient have been demonstrated.

(4) The rapid prototyping of 1-3 composites with novel spatial scale and periodicity via a modified lost mold process was demonstrated. Sacrificial plastic molds was made using either punched wax sheets or polyester microtubes.

(5) A solid freeform fabrication technique was used to form composites with 1-3, 2-2, and 3-3 connectivity. Two approaches were demonstrated: a) composite fabrication via polymeric mold design, and b) composite fabrication via Fused Deposition of Ceramics (FDC<sup>TM</sup>).

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***26 March 1996***

***Afternoon  
Poster Presentations***

## THIN FILMS OF EQUIATOMIC TITANIUM-NICKEL - I: SPUTTER DEPOSITION, MECHANICAL PROPERTIES, AND APPLICATIONS

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Equiatomic titanium-nickel, or 'nitinol', first discovered by Naval researchers in the early sixties, forms the basis of a technically important class of shape-memory and superelastic alloys which have been extensively studied in conventional melt-solidified form. More recently, considerable interest has developed in sputtered thin films of TiNi, and robust shape-memory and superelasticity effects have been demonstrated in thin film materials that show characteristics which are often superior to those that can be achieved in ingot-metallurgy alloys. The unique constitutive behavior of these 'thermotractive' thin films may therefore lead to enabling materials technologies for application to acoustic transducers, smart material systems, damping, fatigue, joining of dissimilar materials, and to microelectromechanical actuators. For example:

- Ohmic electrical excitation can alter the compliance of the material by a factor of two to four, offering the possibility of structural components having controllable mechanical or acoustic impedance, potentially applicable to sensors and transducers.
- Electrical excitation can also radically alter the internal friction and damping capacity of the material. Damping capacity in the martensitic phase (at lower temperatures) is extremely high.
- The martensite-to-austenite transformation can generate large displacements and very high force output: the equivalent of one gram of force can be generated from a 10  $\mu\text{m}$  wide film that is only 2  $\mu\text{m}$  thick. This is several orders of magnitude greater than can be achieved using other available microactuator materials. Applications requiring microminiature actuation, component fastening, structural tensioning, element positioning, or adaptive shaping, may be possible.
- Thin films may be processed to achieve classic transformational superelasticity, as shown in Fig.1. This effect allows anelastic strain excursions to well over 5%, at low stress, while generating few dislocations or other damage artifacts. The strain may be completely recovered upon unloading. This characteristic may, for example, find use in the design of functionally graded interfaces for joining materials having widely different thermal expansion coefficients. It may also be applicable to certain problems in low-cycle fatigue.
- Patterning techniques for titanium-nickel films have been shown to be compatible with microelectronic lithographic procedures, and the films have been successfully deposited and micromachined on polyimide substrates. New microscale interconnect schemes and passive/active dimensional adjustment capabilities may thus be possible.

The present paper will review our recent work on sputter deposition techniques for titanium-nickel films, showing the range of microstructures which can be achieved, and their resultant mechanical properties. Austenite grain sizes below 100 nm have been achieved, giving very high strength and excellent thermotractive properties. Preliminary results have also shown that superelastic TiNi thin films can have a beneficial effect on low-cycle fatigue crack initiation. We will also present results on micromachined, electrically excitable actuators deposited directly onto Kapton polyimide substrates.

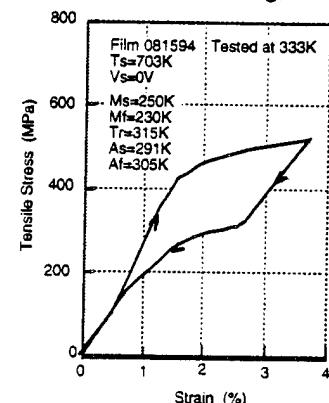


Figure 1. Superelastic Deformation in a Titanium-Nickel Thin Film.

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## THIN FILMS OF EQUIATOMIC TITANIUM-NICKEL - II: MODELING THERMOTRACTIVE TRANSFORMATIONS TO PREDICT THERMOMECHANICAL AND HYSTERESIS RESPONSE

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Development of superelastic and shape-memory alloy thin films for specialized force-producing elements in microelectromechanical systems (MEMS) is motivated by a potential for production of high forces and large displacements in a way which scales well down to the dimensions envisioned for MEMS. Additional aspects of the constitutive behavior of this material system, such as adjustable damping capacity and elastic compliance, may make them useful in acoustic transducer and sensor applications. We have fabricated very robust sputtered thin films of near-equiatomic titanium-nickel that display all of the phase transformations underlying these unique characteristics. In order to predict detailed stress/temperature behavior under cyclic-heating and convective-cooling in thin film systems, and to account for dissipative losses due to transformational hysteresis, we have developed a mathematical model for thermottractive response at a level of detail that permits device modeling and design validation without having to track localized events at the microscale. The model augments conventional continuum mechanical descriptions with internal variables that track fractional partitioning of the microstructure between austenite and the various martensite variants. A three-species model involving austenite and two complementary martensite variants is able to capture the self-accommodated martensite microstructures that support shape memory, and the strain-accommodating structures associated with superelasticity.

Transformations between the different crystal species are tracked on the basis of a nucleation algorithm that reflects the observed hysteresis both for martensite-to-austenite transformation and for martensite twinning-detwinning. This hysteresis is treated by a Duhem-Madelung model that takes complete transformation behavior as constitutive input, and on this basis formally predicts the hysteresis associated with transformation arrest and reversal. Thermodynamic requirements, such as the Clausius-Clapeyron relation, are naturally satisfied in the treatment. A simple algorithm for temperature-dependent uniaxial response requires only experimentally determined parameters: the four transformation temperatures  $M_f$ ,  $M_s$ ,  $A_s$ ,  $A_f$ , the crystallographic transformation strain, the Young's moduli for the austenite and martensite phases, the transformation latent heat, and the triggering and ultimate stresses for martensite detwinning. As such the model captures strain augmentation due to both elastic and transformational response. Specification of material specific heat and convective heat transfer coefficients allows for treatment of heat flow effects both passively (as output) or actively (as input), thus allowing for the simulation of a variety of thermal environments (Figure 1).

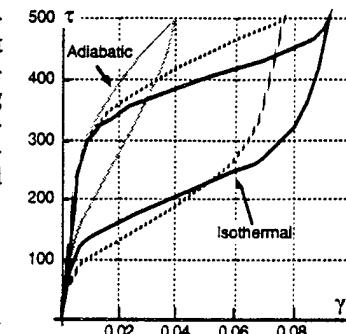


Figure 1. Uniaxial stress-strain simulation for TiNi. Isothermal conditions are optimal for superelastic response, whereas retained heat in an adiabatic environment suppress the transformation. Convective environments (center) give an intermediate response.

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## **DESIGN AND FABRICATION OF PIEZOCOMPOSITE SMART PANELS FOR ACTIVE SURFACE CONTROL**

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Piezocomposite smart panels, capable of both sensing and actuation for active surface control, are currently being developed at MSI. A new approach utilizing monolithic sensor elements built into a 1-3 composite actuator panel offers improved performance over previous designs. The sensors are net-shape formed, low profile accelerometers, uniformly distributed across the panel area. Several accelerometer designs have been modeled and fabricated. Prototype smart panels, integrating sensing and actuation, have been assembled and are being evaluated. Initial test results, focusing on accelerometer sensitivity, component cross-talk, and response uniformity of the active surface, are presented.

This work is supported by the Office of Naval Research and the Advanced Research Projects Agency

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## Materials for Integrated Sensor/Actuator Combinations

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Vibration suppression using active feedback is usually performed with separate sensors and actuators. We will describe an Integrated Sensor/Actuator device shown to be useful for vibration control applications. The active material used was a piezoelectric composite with volume loading of 75%PZT & 25%epoxy.

The evaluations were conducted in a vibration system in which a cantilevered aluminum beam was used to model a component of a large flexible structure. The experimental results showed that the composite had higher voltage sensitivity than bulk PZT when used as sensor in the vibration system, while actuator performance similar to PZT could be achieved through higher feedback gain in the control system.

The experimental results on the integrated sensor/actuator also showed that the composite had good combined sensor/actuator functions in the vibration control system. The performance of the combined sensor/actuator was found to depend on the geometric arrangement of the sensor and actuator. The damping mechanism of the actively controlled vibration system under active negative feedback was attributed to changes in the damping characteristics rather than to changes in the stiffness of the beam.

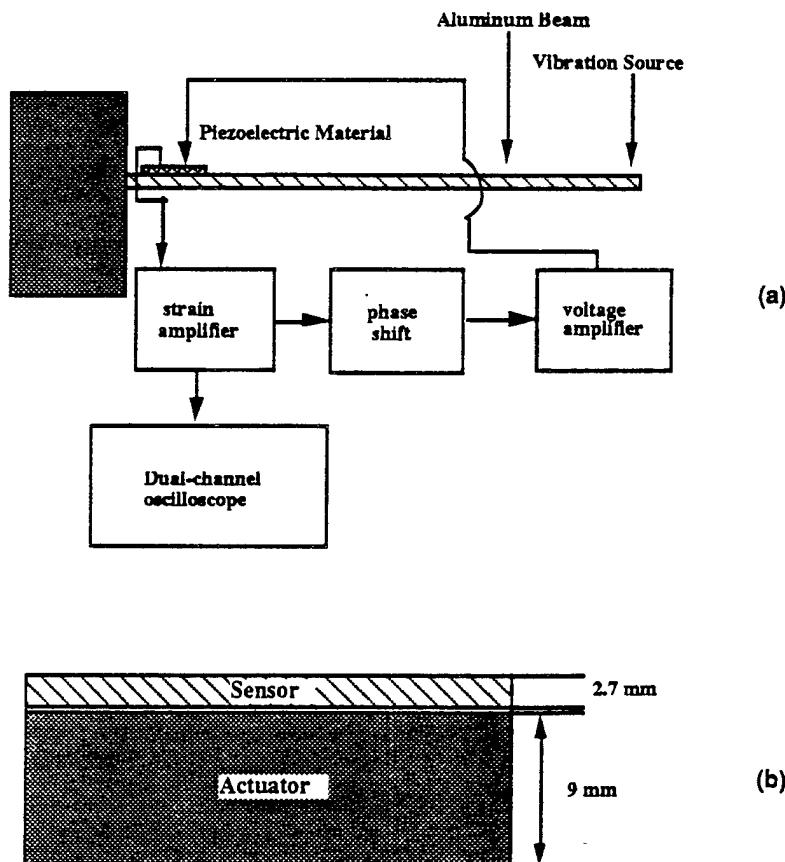


Figure 1 (a) The experimental set-up for vibration control experiment  
(b) Electrode pattern for the integrated sensor/actuator device.

## ANTIVIBRATION SYSTEM WITH PIEZOELECTRIC POLYMER ACTUATORS

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The work described is our first step toward a system meeting NASA requirements for microgravity environments for experiments in space vehicles, and is also aimed at terrestrial applications. In this first step, we are employing feedforward control. We double-integrate the signal from an accelerometer on the shaker simulating the space vehicle. This provides a signal proportional to the shaker position. A high-voltage amplifier sends this amplified signal to the actuator. This signal is intended to provide an actuator displacement equal and opposite to the shaker displacement, thus cancelling any displacement transmitted to the load mounted on the actuator. We will analyze any deviations from this ideal cancellation and try to minimize them, but eventually we expect to employ a feedback control system to provide better cancellation.

The actuator design is based on piezoelectric polymer bimorphs glued together with a double-S precurvature, and specially electroded with narrow electrode gaps at 1/4 and 3/4 of the distance along the stretch direction. Applying a field to the middle half, and an opposite field to the two ends, accentuates or diminishes the curvature, depending on polarity. The basic actuator consists of two such double-S bimorphs glued together at the right and left ends, into a bellows shape. It is then attached at the bottom to the shaker and at the top to the load. The actuator is made of 28 micron PVDF sheets from AMP, with sheet dimensions 3.4 cm wide by 5 cm in the stretch direction. Actuator displacement is 1 mm for a field near 30 V/micron, and maximum force is near 0.2 N.

To "eliminate" gravitational force on a load for simulating space conditions in our lab, we have designed and built a system in which the load is acted on by the equivalent of two springs. One is a normal spring of spring constant  $k$ . The other has spring constant  $-k$ . This is achieved by an axle-jack-like 4-link mechanism. A horizontal rod goes through the left and right bearing pins. It retains compression springs which push inward at both these pins. The springs are adjusted so that these springs exert zero force when the mechanism is fully extended upward. Although these spring forces vanish in this limit, the mechanical advantage becomes infinite in such a way that the effective force increases linearly with displacement.

Work supported by NASA Grant NCCW-0058.

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## LARGE AREA PIEZOELECTRIC COMPOSITE ARRAYS

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Lead zirconate-titanate (PZT)/epoxy composite hydrophones with 3-3 connectivity have been fabricated by embedding reticulated PZT ceramics in an epoxy matrix. Previous work has shown that the hydrostatic properties of these composites can be improved by distorting the ceramic structure from a nearly isotropic network of PZT ligaments toward a highly oriented, laterally reinforced 1-3 configuration. Composites fabricated with Spurr epoxy exhibit good pressure stability and a moderate hydrostatic figure of merit ( $d_{hgh}$ ) of  $\approx 1000 \times 10^{-15} \text{ m}^2/\text{N}$ . The composite elements have been incorporated into a conformable array which displays similar performance (Fig. 1). Techniques have been developed which allow incorporation of the composite into an array configuration which is easily scaleable to production levels and/or large area panels. In addition, other composite configurations that are more sensitive but less pressure stable can be used as array elements so that performance may be tailored to a particular application.

(This work is sponsored by the Office of Naval Research, Grant No. N00014 -92-J-4025)

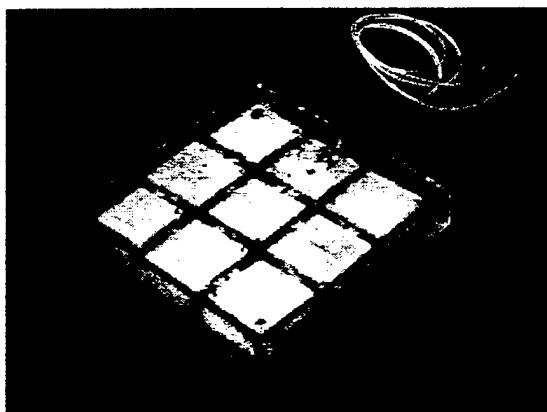


Fig. 1 Flexible, reticulated ceramic composite array (dimensions: 5.7 cm x 5.7 cm x 0.8 cm).

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## PRODUCTION OF DISTORTED 3-3 HYDROPHONE COMPOSITES FROM RETICULATED CERAMICS

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Joint development efforts between Hi-Tech Ceramics and researchers at the New York State College of Ceramics at Alfred University have resulted in a 3-3 PZT/epoxy composite that is light weight, flexible and exhibits moderate hydrostatic response. The fabrication of arrays for testing by the Navy is underway and expected to be completed by 6/96. The manufacturing processes used in the fabrication of these composites are low cost and should result in cost effective devices.

The PZT reticulated ceramic process begins with an open cell polyurethane foam which is distorted by thermomechanical means until the cell aspect ratio is 3:1 or greater. The foam is then cut to the desired dimensions and coated with PZT slurry in one or more deposition steps. The unfired PZT coated foam is fired to burn out the polyurethane and sinter the ceramic. The resulting ceramic has an anisotropic cellular structure and a bulk density of 15-20%. A 3-3 composite is made by infiltrating the PZT reticulate with epoxy, electroding the faces and then poling the structure. The final array is comprised of several of these composites electroded together and joined by a flexible urethane. The size, shape and density of the composite component can be varied depending on the desired end use application.

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\* This is an Industrial Display Poster

## ELECTROSTRICTION IN POLYURETHANES - MORPHOLOGY DEPENDENCE

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Electrostriction measurements on polyurethanes of different morphologies are currently being investigated by our laboratory. Our most recent investigations are on phase mixed systems which have no hard segment domains and yield films of low shear modulus. The phase mixed morphology results in a high value for the  $d_{33}$  coefficient. Values as high as 40A/V (250 Hz) have been observed. This response was measured at an applied bias field of 350V/mil, indicating this material may be a desirable candidate for sonar transducers. These measurements were taken using a dual beam vibrometer which simultaneously measures the displacement on both sides of the polyurethane film. For the measurements reported here the samples were mounted under slight tension and this mounting constraint affects the value of the coupling constant. Further description of the apparatus and sample characterization will be presented.

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1996 ONR TRANSDUCER MATERIALS AND TRANSDUCER WORKSHOP  
ABSTRACT

**MESOSCOPIC INSTABILITY IN TERFENOL-D FILMS**

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Terfenol-D films sputter-deposited onto Si/SiO<sub>2</sub> substrates are in a state of compression, about -500MPa. They display a periodic one-dimensional domain structure which extends over areas of the order of cm<sup>2</sup>. This structure is the result of the large magneto elastic energy of this material resulting in an equally large elastic contribution to the domain wall energy. Due to the compatibility of the strain each domain wall is an elastically distorted layer imbedded between two adjacent domains. Tensioned nanocrystalline Terfenol-D films display a pronounced damping maximum at a magnetic field of about  $\pm 1.5$ kOe oriented perpendicular to the plane of the film. The phenomenon is critically dependent on the orientation of the magnetic field. The composite damping reaches values larger than  $Q^{-1} \approx 0.015$ . These maxima can be retraced applying in- or decreasing magnetic fields and show no signs of hysteresis. When scaled with the present film to substrate thickness ratio a peak value of the film damping of  $Q^{-1} \approx 1$  results. This damping is substantially controlled by a balance of the magneto-elastic and magneto-static energies of the film and reflects an instability of the domain structure in a transverse magnetic field from a periodic polydomain structure single domain structure in which the magnetization is directed perpendicular to the plane of the film. Since the stress in Terfenol-D films sputter-deposited onto Si/SiO<sub>2</sub> substrates can be engineered, so can the domain structure and hence the instability which can be exploited in sensors.

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## MAGNETOSTRICTION OF TERFENOL-D SINGLE CRYSTALS

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Saturation strains greater than 2200 ppm have been measured in fields  $\geq 100$  kA/m in single crystal Terfenol-D ( $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ ) at room temperature. Hysteresis losses were found to be very small. Crystals for this study were grown at the Materials Preparation Center (Ames, IA) in yttria crucibles prepared by spraying yttria powder onto graphite mandrels. The crucibles containing arc-melted melted Terfenol-D were heated in a furnace backfilled with argon to 1500°C (approx. 150°C above the Terfenol-D melting point), and then slowly removed from the furnace hot zone at a constant velocity of 1 cm/hr. Crystals prepared in this manner are *twin-free* and *oriented along the preferred [111] axis*. Saturation strains measured on a 0.46 cm long x 0.8 cm dia. sample are shown in Fig. 1. The insert depicts a typical magnetostriction vs. magnetic field curve measured at an applied stress of 21.2 MPa. Saturation of the magnetostriction for fields  $\geq 100$  kA/m (1.25 kOe) is clearly shown in the insert. The existence of saturation at these fields confirm that the internal losses are low (much smaller than those found in conventional prepared Terfenol-D). These lower losses can be attributed to the absence of [111] twin planes in the single crystals, characteristic of samples prepared at lower temperature gradients. Troublesome Widmanstätten  $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_3$  precipitates were non-existent in alloys containing small excesses of  $\text{Tb}_{0.3}\text{Dy}_{0.7}$ .

We conclude that the use of spray formed yttria crucibles allowed growth rates to be reduced sufficiently to allow for a planar growth front during solidification. With this stable planar interface, undesirable [112] growth is suppressed in favor of preferred twin-free [111] growth.

Large magnetostrictive materials prepared in this manner may be used alone or with high-strain electrostrictive ceramics in hybrid transducers.

*This work is supported by the ONR Acoustic Transduction Technology Task at NUWC and the ILIR Program at NSWC, Carderock.*

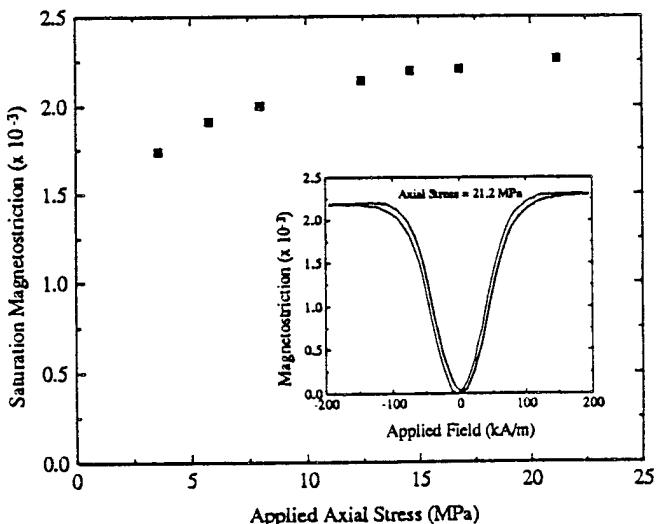


Figure 1. Saturation magnetostriction for single crystal Terfenol-D vs. applied axial stress. Inset shows the magnetostriction vs. field for single crystal Terfenol-D with an applied axial stress of 21.2 MPa.

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# Improved Piezoelectric Ceramic-Polymer Composites for Hydrophones Applications

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Composites of piezoelectric ceramic powders and polymers are described that provide highly desirable properties for sonar hydrophones: (1) high figures of merit,  $g_h d_h = 50(\pm 5) \times 10^{-13} \text{ m}^2/\text{N}$  and  $g_h d_h / \tan \delta = 3.3 \times 10^{-10} \text{ m}^2/\text{N}$ , (2) convenient melt processibility, (3) no significant sensitivity changes in going to high pressures, (4) thermal stability of the poled state for months at 100°C, (5) and high dielectric constants ( $\epsilon = 60-70$ ). These characteristics are reproducibly obtained even for fabricated hydrophones having wall thicknesses of well over a millimeter and cylindrical tube geometries. Such superior performance results from the following materials design choices: *First*, the ceramics and polymer matrix are chosen so that the ratio of polymer dielectric constant to ceramic dielectric constant is unusually high. This choice results in easily poled composites having high remnant polarizations. *Second*, the ceramic powder has a narrow distribution of particle sizes. This choice eliminates the "cage effect" that is inherent in composites prepared from powders having a polydisperse particle size distribution. Such cage effect provides inefficient stress transfer to small particles that reside in cages formed from larger particles. *Third*, the powders utilized have an average particle size that is larger than 40  $\mu\text{m}$ . The consequence of using such large particle sizes is much greater at a ceramic loading level of 50 volume percent than it is at a loading level of 65 volume percent. *Fourth*, an "indicator disk method" is used to insure that ceramic powders used for composite preparation are thoroughly sintered within the individual powder particles, but not between particles. This eliminates properties degradation due to inhomogeneous particle agglomeration. *Finally*, high temperature and high pressure (above 20,000 psi) composite processing effectively removes cavities that could degrade hydrophone properties by producing a pressure dependent sensitivity. Examples of polymer matrices and ceramic powders used for these composites are the  $\alpha$ -phase of poly(vinylidene fluoride) and calcium-modified lead titanate, respectively. Depending upon the piezoelectric stress component that should be optimized for a particular sensor application, the above strategy of sensor composite design can be usefully applied to a host of other piezoelectric ceramics and polymers.

**27 March 1996**

***Morning  
Presentations***

PIEZOELECTRIC CERAMICS FROM  
THE ROSTOV STATE UNIVERSITY, RUSSIA

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**ABSTRACT**

The Department of Active Materials at the University of Rostov at Rostov, Russia maintains a substantial activity in, among others, the development of piezoelectric ceramics. They have a significant number of pertinent publications, including a 30 page description and catalog of more than 50 piezoelectric ceramic formulations.

We have received and are testing electroded discs of 10 compositions and are finding a good correlation to their published values. We are finding some of these to be most competitive with piezoelectric materials that have been available here, ranging from doped lead titanate with  $d_{33}$  values above 85 pC/N to soft PLZT with  $d_{33}$  values above 1000. We also have samples of a (low  $d_{33}$ ) material, said to be operable above 900°C. Where we could find similar U. S. materials, a comparison shows about  $1/2$  of the Rostov materials to have higher  $Q_m$  values, with three out of five having at the same time similar or higher  $d$  parameters. Discussion with Rostov personnel indicated hot pressing to be one of their consolidation methods.

The proposed presentation will include results from this evaluation and from an ongoing composition review.

## Comparison of Russian to Western Piezoelectric Materials

Table A

composition	K*				d33*				Qm				d33 measured				
	PCR number	measured			catalog	measured			catalog	measured			catalog	measured			
		nominal Tc (°C)	Rostov max	NRL 100Hz min		Rostov max	NRL 100Hz min	NRL 100Hz average		Rostov max	NRL 100Hz min	NRL 100Hz average		Rostov max	NRL 100Hz min	NRL 100Hz average	
High temperature	61	1200	50	55	55	57				4.50	<100	12	11	9	14		
PbNb2O6 (1)	570	225				1					11		8.5				
High anisotropy	70+	300	135	135	110	131				1.9	1.19	0.92	1.00	6	110	93	85
Acc. Doped PbCaTiO2 (1)	260	200												62.8		89	
High sensitivity	37+	345	1400	1080	930	994				1.6	4.66	2.43	3.37	105	375	307	
Pb.998(Zr.53 Ti.47)O3 (1)	386	730								0.4				500	223		
Hard PZT	8	325	1400	1545	1195	1450				0.35	0.42	0.38	0.52	2000	290	298	
PZT 8M (2)	305	1050								0.4				1000	295		
HTD†	78	350	1250	1490	1450	1506				0.3	0.29	0.21	0.38	1000	315	297	
PZT 4D (2, 3)	320	1450								0.4				600			
Freq. Stable 0	28	325	600	680	630	703				0.5				2000			
PZT 6B 0	350	460								0.9				1300	71		
Freq. Stable 0	80	310	800	930	760	777				0.6				4000			
PZT 6A 0	335	1050								2				450	189		
Medium Hard	6	230	2300	2350	1580	2111				0.4	0.77	0.44	0.63	300	440	468	
PZT 4S-3 (2)		1490								0.5				260	370		
High K	7M	175	5000	4980	4630	5595				2	2.30	1.79	1.67	60	760	1043	
PZT 5H (2)	193	3400								2					75	593	
High K Lower Tc	73	155	6000	6260	5470	6531				2.9	3.02	2.77	2.60	35	860	1006	

All PCR coded materials are specified and supplied by The Rostov State University.

1) From H. Banno, "Piezoelectric Transducers and Ceramics", Encyclopedia of Advanced Ceramics, 2017-2023, Pergamon, 1994.

2) Private Communication, Morgan Matroc, Bedford, Ohio.

3) PZT 8D has a higher d33 (315 pC/N), but a lower Tc.

For additional footnotes see Table B.

# Comparison of Russian to Western Piezoelectric Materials

Table B

composition description	-d31 measured				-d31/d33 from measured				g33 from measured				pair comparison			
	PCR number	catalog max	Rostov NRL 100Hz min	NRL 100Hz average	catalog max	Rostov NRL 100Hz min	NRL 100Hz average	catalog max	Rostov NRL 100Hz min	NRL 100Hz average	catalog max	Rostov NRL 100Hz min	NRL 100Hz average	catalog max	Rostov NRL 100Hz min	NRL 100Hz average
High temperature	61	0	4	0	0	0.11	0.28	0.11	27.1	28.4	1.06	10.1	max 900 °C operation			
PbNb2O6 (1)		9							43.1				lower operating temp			
High anisotropy	70	0	0	0	0	0	0	0	83	96	0.98	19.9	higher Tc, d33, g33			
Acc. Doped PbCaTiO2 (1)		0							35.3							
High sensitivity	37	170	121	112	127	0.45	0.41	0.42	30.3	35.6	34.9	1.42	19.9	better d33		
Pb.988(Zr.53 Ti.47)O3 (1)		93.5							34.5				better Q, df			
Hard PZT	8	130	124	111	131	0.45	0.44	0.37	31.8							
PZT 8M (2)		110								23.3			better Q			
HTD†	78	130	138	127	135	0.45	0.45	0.43					better g33			
PZT 4D (2, 3)		135								24.6			better Q, d33			
Freq. Stable Ø	28	36	47			0.38				17.3			better Q, d33			
PZT 6B Ø		27								20.4			much better Q, improved d33			
Freq. Stable Ø	80	51	55			0.42							better d33, Q			
PZT 6A Ø		80											better d33, Q			
Medium Hard	6	195	193	144	191	0.44	0.41	0.43		21.6	25.2	1.04	19.9			
PZT 4S-3 (2)		159								28						
High K	7M	350	378	346	442	0.46	0.42			17.2	21.1	1.14	25.0			
PZT 5H (2)		274								19.7			lower k, d33			
High K Lower Tc	73	380	375	357	445	0.44	0.44			16.2	17.4	1.04	20.0			

All measurements at 25°C

All d parameters in pC/N

All g33 parameters in 10^-3 Vm/N

\*  $\mathcal{E}_{33}/\mathcal{E}_0$

\*\* tg δ × 10^2

d31 = d31(measured) × thickness/dia

† Higher Temperature Driver

+ Monel Electrodes: (4Ω lateral resistance with 1 cm spacing)

69% Ni

1% Fe

29% Cu

1% Mn

(all other samples have Silver Electrodes)

◊ δf0/fr % (-60 °C to +85 °C) < 0.25%

## FIELD INDUCED ANTIFERROELECTRIC-TO-FERROELECTRIC PLZST CERAMICS

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It has been over thirty years since the initial, comprehensive papers on modified lead zirconate titanate stannate (PZTS) ceramics by Berlincourt and others<sup>1,2</sup> were published. Since then further investigation of this family of ceramics was provided by Uchino<sup>3</sup>, Cross<sup>4</sup>, and many others. This paper describes our studies on lanthanum modified PZTS ceramics for potential applications such as low frequency sonar projectors and large strain electromechanical actuators.

In the previous ONR review meeting various compositions within the  $Pb_{0.98}La_{0.02}(Zr,Sn,Ti)_3O_3$  (PLZST) ternary phase diagram along with Sr and Ba modified compositions were discussed. This paper describes both additional compositional studies and in-situ XRD research confirming a maximum strain level, in the direction of the applied E-field, as being approximately 0.59%. This value was calculated based on the lattice change from tetragonal antiferroelectric (AFE) to rhombohedral ferroelectric (FE). The PLZST composition x/y/z:55/12/33 was extensively studied in order to compile necessary data with respect to actuator applications. The data included driving requirements (dielectric constant and loss in both high and low fields), operating temperature range, and the effect of frequency. Mechanical properties are also discussed in both the AFE and FE states. The moduli of both the AFE and FE states were in the range of 200GPa. However, the displacement data under pre-stressed conditions using a stress bolt indicated that the phase switching motion was rather soft and involves more than one step. The data is compared to other electro-active piezoelectric and electrostrictive ceramics. Further detailed information will be discussed in two poster presentations.

**Acknowledgement:** This work was performed under ARPA Smart Structure for Rotorcraft Consortium (SSRC). XRD studies were performed in collaboration with C. Hicks and C. Blue of NCCOSC/RDT&E Division.

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***Morning  
Poster Presentations***

## DOMAIN RELATED PHASE TRANSITIONS IN LEAD ZINC NIOBATE RELAXOR FERROELECTRIC SINGLE CRYSTALS

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Relaxor ferroelectric lead zinc niobate,  $Pb(Zn_{1/3}Nb_{2/3})O_3$ , has been intensively investigated, intrigued by its large dielectric and piezoelectric properties which are useful in designing actuators and transducers. The unique characteristics of this material may originate from the domain configuration which is composed of ferroelectric microdomains. In this study, the in-situ behavior of the macrodomains in  $Pb(Zn_{1/3}Nb_{2/3})O_3$  single crystals were observed using an optical microscope in combination with a CCD camera system. This equipment configuration allowed the temperature of the sample to be cycled between -185°C to +200°C, while safely applying an electric field up to  $\pm 10$  kV/cm. Many characteristics such as induction of birefringence, transition between the microdomains to macrodomains, and "freeze-in" temperature of the macrodomains were optically observed. These optically observed characteristics were then compared to measured dielectric properties in an attempt to clarify the relationship between dielectric properties and macrodomains. The optical and dielectric data were collected and plotted as a function of temperature and electric field. A phase diagram divided into four domain related regions is proposed.

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CRYSTAL GROWTH AND FERROELECTRIC RELATED PROPERTIES  
OF (1-x)  $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$  - x  $\text{PbTiO}_3$ <sup>†</sup>

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Crystals of (1-x)  $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$  - x  $\text{PbTiO}_3$  were grown using the flux technique. The effect of growth variables such as: flux to composition ratio, soaking temperature, and cooling rate on crystal quality were investigated. Variations in stoichiometry of crystals and associated variations on dielectric properties due to the growth conditions will be presented. Two crystallographic axes—the principal symmetric and the pseudocubic axis—were employed to characterize piezoelectric and ferroelectric related properties. When the crystal has rhombohedral symmetry ( $x < 0.9$ ), samples show large differences in piezoelectric properties according to the axis. This could be ascribed not only to its own crystallographic origin, but also to the stability of domain orientations after poling. This will be discussed based on polarization and strain behavior associated with the application of a unipolar electric field.

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# ANTIFERROELECTRIC-TO-FERROELECTRIC PHASE SWITCHING PLZST CERAMICS- I. STRUCTURE, COMPOSITIONAL MODIFICATION AND ELECTRIC PROPERTIES

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Electric field induced antiferroelectric (AFE) to ferroelectric (FE) phase transformations are accompanied by large strain and significant hysteresis. The properties of these materials can be tailored to fit specific applications such as high strain actuators and charge storage capacitors.

In this study the phase transformation behavior of antiferroelectric  $(\text{Pb}_{0.98-\delta}\text{La}_{0.02}\text{A}_\delta)(\text{Zr}_x\text{Sn}_y\text{Ti}_z)\text{O}_3$  ( $\text{A}=\text{Ba, Sr}$ ) ceramics have been investigated. First, in-situ x-ray diffraction studies were conducted on a representative composition to identify the crystal structure of the phases. From this data the maximum theoretical strain associated with the AFE-FE transformation could be calculated. Compositional modifications on both the A and B sites were attempted in order to modify the material properties. The B-site modifications were completed through manipulation of the Ti:Sn and Zr:Sn ratios. Decreasing the Ti:Sn ratio produced increases in the switching field, decreases in AFE-FE transition temperature, more diffused dielectric maximum peak and decreases in the room temperature dielectric constant. Regardless of the Ti:Sn ratio, 0.2% strain was realized at the switching field and hysteresis remained constant. On the other hand increases in the Zr:Sn ratio produced lower switching field, increased hysteresis and increased AFE-FE transition temperature, the dielectric maximum increased and the dielectric maximum peak became sharper. A-site modifications were accomplished by the addition of Ba and Sr. Ba proved to be a strong FE stabilizer while Sr proved to be a strong AFE stabilizer.

Processing issues including an evaluation of the advantages of HIPping as well as the effect of increases in sintering temperature up to 1400°C will be addressed. Some practical data including current requirements and heat generation will also be presented.

**Acknowledgement:** This work was performed under ARPA Smart Structure for Rotorcraft Consortium (SSRC). XRD studies were performed in collaboration with C. Hicks and C. Blue of NCCOSC/RDT&E Division.

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## ANTIFERROELECTRIC-TO-FERROELECTRIC PHASE SWITCHING PLZTS CERAMICS- II. THE EFFECT OF PRE-STRESS CONDITIONS ON THE STRAIN BEHAVIOR

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Actuators used for large strain applications are typically placed in pre-stressed conditions to maintain their integrity during service. There is a lack of research available regarding the study of antiferroelectric (AFE) materials under pre-stressed conditions and the subsequent effect on their properties. Past research typically involved PZT based materials but the analysis concentrated on the effect on piezoelectric coefficients and dielectric properties, which were determined primarily using resonance methods and low fields. Conversely, studies involving AFE phase change materials require the application of high fields and analysis of changes in strain.

In this study the effects of pre-stresses in  $(\text{Pb}_{0.98-\delta}\text{La}_{0.02})(\text{Zr}_x\text{S}_{1-x}\text{Ti}_z)\text{O}_3$  (PLZST), hard and soft PZT and PMN were examined, with an emphasis on the phase transformation behavior of PLZST. The polarization and displacement of the samples were measured under stress-free as well as various pre-stressed conditions. It was found that the performance of PLZST is very sensitive to pre-stresses along the electric field direction. Specifically, the displacement decreased by 50% from its stress-free value as soon as a small pre-stress (3MPa) was applied. This phenomenon can be explained by the switching mechanism in this system, based on the X-ray diffraction data in Part I and the polarization and strain behaviors of the material. The results showed that the large decrease in displacement is mostly due to the suppressed (soft) piezoelectric activity after AFE->FE transformation. This observation is compared to the behavior of other electro-active ceramics.

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# In-situ x-ray diffraction study of the antiferroelectric-ferroelectric phase transition in PLSnZT

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In-situ x-ray diffraction studies were performed on the PLSnZT antiferroelectric-ferroelectric phase switching ceramic and polycrystalline powder. The crystallography of both the antiferroelectric and electric field-induced ferroelectric phases were determined for the approximate composition of  $Pb_{.98}La_{.02}(Zr_{.55}Sn_{.33}Ti_{.12})_{.995}O_3$ . A preferred antiferroelectric domain structure has been identified and possible mechanisms responsible for the domain structure presented. A single tetragonal phase has been identified for the room temperature zero-field antiferroelectric material with unit cell dimensions  $a=4.107\text{\AA}$  and  $c=4.081\text{\AA}$ . An electric field-induced structure developing at the antiferroelectric-ferroelectric switching field has also been observed and determined to be of rhombohedral symmetry with  $a=4.105\text{\AA}$  and  $\theta = 89.85^\circ$  indicating a volume increase of 0.49% for the tetragonal-rhombohedral transition.

## THE EFFECT OF ANNEALING TEMPERATURE ON THE FORMATION OF $\text{SrBi}_2\text{Ta}_2\text{O}_9$ (SBT) THIN FILMS

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In this paper we report on synthesis of SBT thin films by sol-gel processing. Sr metal, Bi-2 ethyl hexanoate and Ta-ethoxide were used as precursors. Thin films with nominal compositions  $\text{SrBi}_2\text{Ta}_2\text{O}_9$  and SBT +10% excess Bi content were made. Films were annealed at various temperatures to study the microstructure, crystallization temperature and the polarization values. Good crystallization of SBT was obtained by annealing at  $700^\circ\text{C}$  - 2 hrs, independent of the Bi content in the films. Films annealed in oxygen atmosphere at  $800^\circ\text{C}$  - 2 hrs did not show any significant change in the polarization value. Crack free films were made with film thicknesses of  $0.4 \mu\text{m}$ . Films annealed at  $800^\circ\text{C}$  - 2 hrs showed a grain size of  $\sim 0.2 \mu\text{m}$ , and reasonably good polarization values of  $5 \mu\text{C}/\text{cm}^2$ . In contrast, films prepared with 10% excess Bi showed a very fine grain size  $< 0.1 \mu\text{m}$  with a lower polarization values of  $1.5 \mu\text{C}/\text{cm}^2$ .

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## THIN FILM ACTUATOR MATERIALS

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Ferroelectric and antiferroelectric thin films are candidates for actuators in microelectromechanical systems (MEMS). However, thin films differ from bulk actuator materials in several significant ways: the grain size is usually smaller ( $<\sim 0.5\mu\text{m}$ ), films are typically under in-plane stress due to either the deposition or to substrate/film thermal expansion induced mismatch, and the breakdown strengths are appreciably higher. The first two factors can lead to considerable degradation in the properties of thin films, which can partially be recovered by the ability to drive devices at higher field levels or under bias.

In this work, electrical and electromechanical measurements were made on a series of ferroic thin films of interest for actuators. In particular, undoped lead zirconate titanate (PZT) prepared by sol-gel and by sputtering, sputtered  $\text{PbZrO}_3$ , and laser ablated PZT-5A films were examined. In all cases, the films were well-crystallized. Well-defined double hysteresis loops (maximum polarization of  $40 - 70\mu\text{C}/\text{cm}^2$ ) and elevated transition temperatures were obtained for the sputtered  $\text{PbZrO}_3$  films. Similarly, good hysteresis loops were obtained on the ferroelectric films. These properties are currently being investigated as a function of the applied in-plane stress, measured by loading the sample in biaxial flexure. This should lead to insight on the importance of stress in modulating the properties of thin films. The piezoelectric (or phase-change induced strain change) properties of each of these films will be reported.

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# MOLECULAR DYNAMICS SIMULATIONS OF PMN CERAMICS

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The future Navy emphasis for improved and affordable high-power sonar arrays and transducers capable of performing in shallow water environments is driving the demand for novel transducer materials capable of satisfying the conflicting requirements of high radiated power, light weight, and low-cost. Electrostrictive PMN relaxor ceramic has emerged as a potential breakthrough material for future sonar transducers. Its huge dielectric constant, large strain, low electro-mechanical hysteresis, and high energy density make this material an attractive candidate for low cost sonar designs relying on low weight drivers using the same or less power as conventional PZT ceramics.

In recent years, molecular dynamics has emerged as a promising methodology to probe structure-property relationships on solid materials, that is to develop an *a priori* understanding of the properties of materials. We have performed simulations on PMN crystal structures with and without lanthanum substitution in the A-site. These preliminary calculations have given us sufficient confidence in molecular dynamics. Using constant-pressure constant-temperature molecular dynamics to simulate 1173 K (900 C) annealing of pure PMN and PMN with lanthanum substitution in the A-site, we discovered a remarkable difference in the self-diffusion constants within these two compositions at this temperature. The self-diffusion constants are about  $10^{-4}$  to  $10^{-3}$  Å/ps/atom in undoped PMN, whereas in lanthanum-doped PMN they are 10 to 100 times larger. It is evident that in undoped PMN, atomic motion is restricted, in contrast to lanthanum-substituted PMN where ionic motion occurs. We propose that at 1173 K (900 C), lanthanum permits increased ionic mobility. This might happen because with lanthanum in the A-site, there is no charge imbalance to "lock" the system in a deep potential energy well. Without electrostatic charge forces restricting ionic mobility, the atoms in lanthanum-substituted PMN undergo considerable diffusive displacements, allowing the system to explore the three-dimensional energy landscape, to surmount the small energy barriers, and eventually to achieve the relaxed state.

The picture emerging from this work strongly supports the well-documented observation that donor doping of PMN with lanthanum results in significant enhancement in ordered domain size, whereas under similar annealing conditions, undoped PMN does not experience any changes in domain size.

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## **THERMOCHEMISTRY AND NON-OHMIC ELECTRICAL CONTACTS IN ELECTROCERAMIC MATERIALS**

**C.A. Randall and D. Cann**

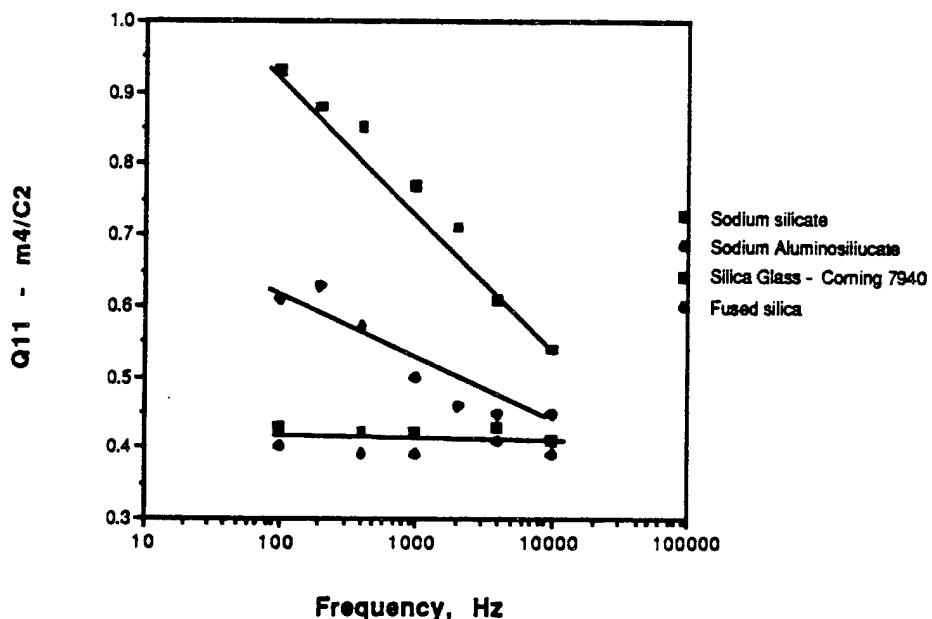
As trends in dimensional control increase and device performance is pushed to higher limits, we generally maximize the processing, doping and microstructure of the electroceramic material. In electroceramic applications, the understanding of the electrode and electroceramic material interface is not as well developed. In this presentation, we will outline some new results which test our hypothesis that the thermochemistry of the interface and electrical characteristics are fundamentally associated with the same interactions. We demonstrate metal-oxygen and metal-metal interactions can be considered in the thermodynamic work of adhesion. In turn, there are corresponding variations to the electronic band structure at the interface. Impedance spectroscopy is used to characterize the variations in contact resistance and space charge capacitance in selected electroceramics with various metallizations. These results are discussed with actuator materials in mind, but we will also allude to the wider implications.

# THE ROLE OF POLARIZATION MECHANISMS IN ELECTROSTRRICTIVE EFFECTS FOR LOW PERMITTIVITY GLASSES AND CERAMICS

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The polarization mechanisms that affect the dielectric and electrostrictive responses of low permittivity dielectrics are examined in this work. The dielectric responses of sodium trisilicate and sodium aluminosilicate glasses in the frequency range 100Hz - 10kHz show a relaxation around 160Hz. This is not a single dipole relaxation. The electrostriction  $Q_{11}$  coefficient of the sodium silicate glass decreases from 0.95 to 0.55  $\text{m}^4/\text{C}^2$ . In contrast, the  $Q_{11}$  of the sodium aluminosilicate glass decreases from 0.6 to 0.45  $\text{m}^4/\text{C}^2$  in the same frequency range. Two samples of vitreous silica show little variation of  $Q_{11}$  with frequency in this range ( $Q_{11} \sim 0.4 \text{ m}^4/\text{C}^2$ ). The expanded coordination shell around the  $\text{Na}^+$  ion in the sodium aluminosilicate glass could cause the more gradual decrease of  $Q_{11}$  with frequency. Results have been reported on markedly different electrostrictive behavior in pure alkali halides and alkali halides doped with low ( $\sim 10^{15}$  ions/cc) concentrations of ions such as  $\text{Li}^+$  and  $\text{OH}^-$ . These different contributions of the host lattice and the defect dipoles for these materials are also analyzed. This work was performed as part of the NSF/MRG project DMR 9223847.

## Electrostriction in Glasses



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## STUDIES ON THE DEVELOPMENT OF PIEZOELECTRIC TRAVELING WAVE MOTORS

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This paper summarizes findings of a joint program between UMR and Allied Signal to develop traveling wave motors (TWMs). Research efforts are focused on issues related to the piezoelectric elements, analytical modeling of the TWMs, and on overall miniaturization of the motor.

Studies on the piezoelectric have been directed towards optimization in terms of its processing (microstructure, strength, surface finish), electrical properties, and resistance to depoling. In this program we are investigating the effect of these issues using both hard and soft PZT formulations. Motor elements have been prepared using tape cast structures, with emphasis on achieving requisite geometries (down to 50  $\mu\text{m}$  thick) without any machining.

An analytical model was also created to help develop a motor with improved performance and durability. The model allows for the selection of many different material properties and design geometries for better performance and reliability, while accounting for manufacturing limitations such as piezoelectric plate thickness and bond stiffness. An analytical model was also developed for the friction force and stick-slip parameters based on the geometry of the motor and the deformations of the stator by combining theories of plate mechanics with those in contact mechanics. This allows the determination of the torque and rotational speed of the motor, and provides a method for finding the effect of using different contact materials in the contact region.

At the poster session several operating small (8mm) motors will be displayed.

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***INDUSTRIAL DISPLAY POSTER***

## COMPACT ULTRASONIC MOTOR

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A compact ultrasonic motor was newly designed and constructed. The motor was designed with the aim of keeping the number of components to a minimum. The components that were used include piezoceramic disc, thrust bearing, rotor and spring. The size of the motor is also small with 10 mm in rotor diameter. The piezoceramic disc is used in the radial mode and excited with two sinusoidal signals 90 degrees out of phase. The inner circle was used for generating the rotation of the rotor. The piezoceramic disc should have a high mechanical quality factor, high coupling coefficient, low dielectric loss and good temperature stability. The piezoceramic disc was excited at the fundamental resonance frequency. ((1,1)) and ((2,1)) modes were configured for excitation. Speed - Torque characteristics were measured for motors with different compositions of piezoceramic discs. Displacement in the rotor was measured using a optical-fiber sensor. Temperature rise due to heat generation was measured. Materials that have low dielectric loss were chosen to reduce the temperature rise. The work was sponsored by *Office of Naval Research*.

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## RAINBOW ACTUATOR STACKS AND ARRAYS

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Previous work on high displacement Rainbow actuators has shown that they possess the capability to be configured into linear stacks for higher displacement devices or into larger area arrays for actuator/sensor functional components. The subject of this investigation was to demonstrate this capability even further by constructing working models of each type, i.e., actuator stacks and arrays. Since it is already known that individual Rainbow pancake actuator elements have the capability of achieving high electromechanical displacements (200 microns) with a unipolar voltage (450 volts) and twice that amount for bipolar operation in a dome mode, the present work focused on linearly stacking several Rainbow elements together in multiple groups of two, as in a clamshell arrangement, and then bonding them together into a single unit. The characteristics of these Rainbow stacks (four clamshell units = a short stack and eight units = a long stack), ranging in diameter from 12.7 mm to 31.75 mm, were evaluated as a function of voltage and load bearing properties. For example, a 12.7 mm dia x 24 mm long PLZT Rainbow long stack (1/53/47) achieved 725 um displacement with no loading and 380 um with 1 kg point load and 180 um with 3.5 kg point load. A similar long stack with dimensions of 31.75 mm diameter and 35 mm long achieved 318 um displacement with a point load of 5.5 kg. Total displacements were found to scale linearly with the number of individual Rainbow units.

Three different Rainbow arrays (9.5x11, 11.5x16, and 14x17 cm) consisting of 42 ea. 12.5 mm dia. elements, 18 ea. 31.75 mm dia. elements and 20 ea. 31.75 mm dia. elements, respectively, were fabricated using lead foil (0.2 mm thick) as top and bottom retaining plates. All of the arrays were one Rainbow element thick leading to an overall thickness of approximately 1.25 mm for each of the arrays. Lead foil was selected as the supporting material because its high ductility allowed a device to be used as a conformal actuator/sensor when applied to either a flat or a curved surface. The arrays were qualitatively evaluated for sensor and actuator functions.

This work was supported by ONR under grant No. N0014-94-1-0563

# STRESS AND FATIGUE ESTIMATION IN MULTI-LAYER CERAMIC ACTUATORS USING AN INTERNAL STRAIN GAUGE

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Nowadays, the multilayer ceramic actuator is a key component in micro-mechatronics. The actuator's reliability and fatigue problems have become an important issue for practical use with an increasing number of applications. In the previous work, crackings and delamination in the multilayer ceramic actuators were observed. In this study, the internal strain in a model actuator with a simple interdigital electrode structure has been investigated. In order to measure the internal strain, an internal strain gauge was employed by printing a gauge shape Pt electrode on a green tape (Fig. 1). The model-actuators were fabricated by a tape-casting method. The internal strain and the electrode fatigue were observed as a resistance change in the internal strain gauge, which was used as a ground side electrode. After poled, the remnant displacement was observed as a decrease in the resistance at 0V. This change corresponds to the remnant transverse displacement. The resistance decreased with applied voltage, because of the transverse displacement. The resistance gradually increased with the number of driving cycle. However, a drastic increase of the resistance with applied voltage, which might be caused by a disconnection of the internal electrode, was observed after the cyclic test. This work was supported by the Office of Naval Research through Contract No. N00014-92-J-1510.

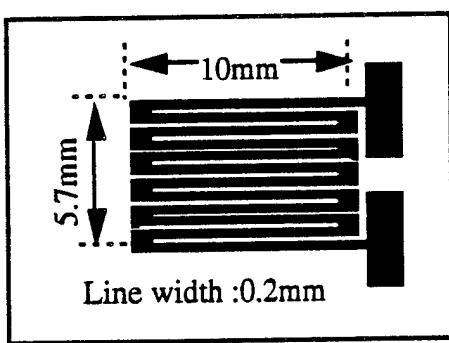


Fig.1 Gauge Electrode

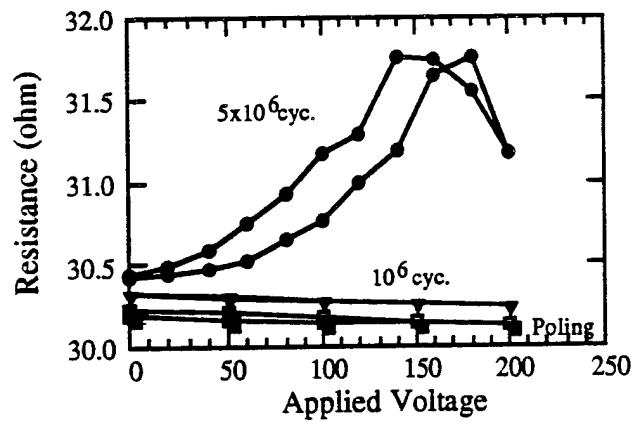


Fig. 2 Resistance change after cyclic test

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# CRACKING IN FERROELECTRIC CERAMIC MULTILAYER ACTUATORS

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## ABSTRACT

Although a ceramic multilayer actuator is normally operated below the coercive field, abrupt ends of internal electrodes concentrate the electric field. Near the internal electrode end, the electric field intensity exceeds the coercive field, which causes local ferroelectric domain switching. The switching induces a large incompatible strain field, which, in turn, produces a high stress field in the ceramic. Crack nucleation and growth have been observed experimentally, but have not been well modeled due to complex material behaviors. In this paper, we idealize, macroscopically, the ferroelectric switching as large increases of both the electric displacement and strains under a small change in the electric field. Ignoring the stress contribution to the electric field, we solve the electric field around the internal electrode edge for various nonlinear electric field and electric displacement relations both analytically and numerically. Taking step-like and quadratic electrostrictive strains as examples of field-induced strains, we solve standard elastic residual strain problems to determine stresses in the structure. In the end, we apply fracture mechanics to determine the cracking conditions in the ferroelectric ceramic multilayer actuators. We find that a critical thickness exists below which cracking will not occur, and analytical solutions based on small-scale saturation conditions are valid under large-scale saturation.

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## CERAMIC-METAL COMPOSITE TRANSDUCERS FOR UNDERWATER ACOUSTIC APPLICATIONS

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A new type of roto-flextensional electroacoustic transducer has been developed for use as a shallow water projector and/or receiver. The transducers currently being studied are 12.7mm in diameter, 2mm thick, and weigh less than 2 grams. They can be utilized at frequencies between 10Hz and approximately 40kHz. Effective piezoelectric voltage coefficients ( $g_h$ ) of these devices have been measured and can range from 20mV•meters/N to nearly 200mV•meters/N, which is 5 to 50 times greater than that of the bulk PZT itself. In addition, the transducers retain the large capacitance of the PZT element, which is between 1500pF and 3000pF.

A hydrophone consisting simply of a bulk piezoelectric (such as PZT) exhibits low sensitivity when completely submerged in water because of its low  $d_h$  ( $=d_{33} + 2d_{31}$ ), high dielectric constant (K), and subsequently low  $g_h$ . By attaching a thin metal cap containing a shallow cavity on its inner surface to each face of an electroded PZT disk, the sensitivity is greatly enhanced. The caps serve to redistribute the hydrostatic pressure in such a way so that a portion of the axial direction stress is transformed into radial and tangential stresses of opposite sign, thereby allowing the  $d_{33}$  and  $d_{31}$  contributions from the PZT to now add in the effective  $d_h$  of the device rather than subtracting. The effective  $g_h$  of the transducer ( $=d_h/K\varepsilon_0$ ), is proportional to its receive sensitivity.

Data obtained experimentally as well as from computer models will be presented to show the effect of PZT type, cap material, dimensions, and geometry on the pressure tolerance, sensitivity, and working frequency range of the transducer. It will be shown that transducers with stiffer and/or thicker endcaps exhibit better pressure tolerance than those with more compliant and/or thinner caps, but at the cost of reduced sensitivity.

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## **BIMORPH BASED DOUBLE AMPLIFIER: A POTENTIAL TRANSDUCER USED IN AIR ACOUSTICS**

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A new type of piezoelectric transducer has been developed for air acoustics and active noise control. The transducer is based on the composite panel structure of bimorph based double amplifier, that is, two parallel bimorphs or bimorph arrays with a curved cover plate as an active diaphragm attached to the top of the bimorphs. The electro-acoustic performance of the transducer depends directly on the displacement of the cover plate and the resonant frequency of the double amplifier structure, which determine the emitted sound pressure level and the working frequency range of the transducer. The displacement of the cover plate and the resonant frequency of the structure depends on the dimensions and materials of the bimorph actuators, the dimensions of the double amplifier structure, and the cover plate materials. In this work, the effects of these factors are investigated systematically, and the optimum double amplifier structure was obtained experimentally. The displacement of the cover plate of the transducer constructed from the double amplifier structure can reach millimeter scale with a relatively low driving voltage, which is more than ten times larger than the tip displacement of bimorphs. The transducer can work effectively below 1000Hz with the sound pressure level more than 90dB (near field) and 80dB (far field), which is suitable for air acoustics applications. Because of its light weight and panel structure, the transducer is especially promising for active noise control as a sound transmitter.

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## BIMORPH-BASED AIR TRANSDUCER: A MODEL

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A new type of bimorph-based air transducer has been recently developed. It consists of two arrays of piezoelectric cantilevers bridged by a curved diaphragm. The purpose of this study was to create a model describing vibration characteristics of this transducer. We suggest one-dimensional approach where the diaphragm is considered to be a rigid one. This diaphragm creates inertia, elastic, and damping forces that affect vibrations of the piezoelectric bimorphs. As a result the resonant frequency of the system changes. To include losses in piezoelectric elements, we use complex piezoelectric, dielectric and elastic constants. Results of modeling and experimental data are in good agreement. This model can be used for optimizing acoustic characteristics of the transducer.

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# THE "MOONIE" AND "CYMBAL" ELECTROMECHANICAL ACTUATORS

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The moonie and cymbal actuators are new types of ceramic-metal composite transducers which are based on the concept of the flextensional and roto-flextensional transducer, respectively. Both the moonie and cymbal actuator consist of a piezoelectric, electrostrictive, or antiferroelectric-ferroelectric phase change material sandwiched between two specially shaped endcaps containing a cavity on their inner surface. These endcaps can be made of metal, polymer, polymer-based composites, or glass based materials.

A brass-capped moonie 12.7mm in diameter and 2mm thick, which utilizes a single layer PZT5 disk 12.7mm in diameter and 1mm thick can exhibit a maximum displacement of 22  $\mu\text{m}$  at 1.0 kV/mm. Using a multilayer ceramic in the moonie design, the applied voltage can be decreased to 100V while retaining an equivalent displacement. The maximum generative force of a moonie actuator with a brass endcap thickness of 0.3mm is approximately 3N at the center of the cap. Both the displacement and generative force show a position dependent behavior, however. The fastest response time of the moonie actuator varies between 5 and 50  $\mu\text{sec}$  depending upon the cavity size.

A new endcap geometry called the "cymbal" has been developed which provides an additional roto-flextensional mode. The cavity of the cymbal endcap has a truncated cone shape. A punch and die were designed to fabricate identical endcaps at minimal cost. Cymbal actuators with identical outside dimensions as the moonie now show displacement values of about 40  $\mu\text{m}$ , with less position dependent behavior. They also have higher generative forces (15 N) due to the enlarged active surface area and the reduced metal content.

Fatigue tests on multilayer moonie actuators with brass caps were performed at room temperature under a cyclic electric field of 1kV/mm with triangular wave form at 100Hz, up to  $10^7$  cycles. Deviations of less than  $\pm 0.1\%$  from the original displacement value were observed. The resonance and antiresonance frequencies as well as their peak amplitudes were the same as the original measured values before the fatigue test which indicates that there is no degradation of the bonding layer. The second essential reliability test is the temperature dependence of displacement. A  $\pm 15\%$  nonpermanent deviation in displacement from room temperature was observed for the brass capped moonies over the temperature range -20°C to +70°C. The thermal expansion coefficient difference between the PZT ceramic driving element and the endcap causes this large thermally induced displacement. Utilizing Kovar endcaps (a Ni-Fe-Co alloy), which has the same thermal expansion coefficient as PZT, eliminates the effect of thermally induced displacement.

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# RESULTS OF 8 mm ULTRASONIC MINIMOTOR DEVELOPMENT USING DESIGN OF EXPERIMENTS

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In an effort to design improved thin-film piezoelectric ultrasonic micromotors on silicon<sup>1</sup> a number of models were proposed for the contact mechanics at the rotor-stator interface and a designed experiment was performed on 8 mm diameter by 3 mm tall motors fashioned from bulk PZT [1]. Results show significant improvements in stall torque density and power density over conventional electromagnetic motors of comparable size and point to new directions for improved microfabrication process and model development.

Two generations of bulk-PZT 8 mm ultrasonic motors were actually fabricated. An interferometer was used to characterize out-of-plane displacements of the stator and a custom-built dynamometer was used to measure torques under 1 mNm in magnitude. First-generation trials on 8 mm motors pointed to a need for an improved bonding technique and a more space-efficient electrode pattern. Both modifications were incorporated into the second generation motors.

The second generation motors were created using Design of Experiments techniques in a one-quarter fraction factorial design. Seven parameters were varied in a binary fashion in the experiment: stator tooth height, stator base height, number of teeth, stator material, rotor material, rotor lining and lubricant. Of the  $2^7$  possible combinations,  $2^5$  trials were actually undertaken, where maximum stall torque and maximum no-load speed were measured for each trial. Optimal parameters for minimizing the stall torque or no-load speed, and a predictor polynomial for any given trial were determined using search and regression analysis. Subsequent to the designed experiment trials, profilometer measurements were performed on the stator and rotor surfaces for characterization of wear. These devices demonstrated maximum stall torques of  $10^{-3}$  mNm, maximum no-load speeds of 1710 rpm and peak power outputs of 27 mW. The resulting peak power density of 108 W/kg is more than double that of human muscle. The maximum stall torque density, found to be 2.9 Nm/kg, is 50 times that of the smallest commercially available component DC motor, which also happens to be 5 times larger and is not sold with a gear. Details of results, comparisons to models and future directions for micromotor development will be discussed.

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<sup>1</sup>Results of first fabrication attempts in a joint project between the MIT Artificial Intelligence Laboratory, the Pennsylvania State University Materials Research Laboratory and Lincoln Laboratory's Solid State Division, were reported in [2]. This latter work on bulk PZT motors was performed at the MIT Artificial Intelligence Laboratory under the ARPA Biomedical Technology Program, contract number N00014-94-1-0069.

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***Afternoon  
Presentations***

## RELIABILITY OF CERAMIC ACTUATORS

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Reliability of ceramic actuators is dependent on complexed factors, which are divided into three major categories: reliability of the ceramic itself, reliability of the device design and drive technique issue. We have been studying the reliability issues from whole points of view.

Investigation on the compositional change of the actuator ceramics and the doping effect was a primary issue to stabilize temperature and external stress dependence of the induced strains. Grain size and porosity control of the ceramics was another key point to realize the reproducibility of the actuator characteristics. Aging phenomenon, i.e. degradation of the strain response, is, in general, strongly dependent on the applied electric field as well as on the circumstances such as temperature, humidity and mechanical bias stress.

The device design affected considerably its durability and life time. Silver electrode metal tends to migrate into the piezoceramic under a high electric field in high humidity. Silver-palladium alloy usage suppressed this problem effectively. Also resistive coating of the device should be taken into account. To overcome the delamination of the electrode, improvement of the adhesion could be realized by using a mesh-type electrode or an electrode material with mixed metal and ceramic (the matrix composition!) powders. Pure ceramic electrode materials were also developed using semiconductive perovskite oxides (barium titanate-based PTCR ceramics).

Particularly in multilayer type actuators, reduction of the tensile stress concentration around the internal electrode edge of the conventional interdigital configuration was the main theme. Three electrode configurations have been proposed: plate-through, interdigital & slit, and interdigital & float electrode types. The last "float electrode" type is a very promising design and will be described in details in the presentation. An empirical rule "the thinner the layer is, the tougher the device is" is also very intriguing, which must be theoretically verified in the near future.

Failure detection or life time prediction methods of ceramic actuators will remarkably increase the reliability against their users. Acoustic emission and surface potential monitoring are promising methods. A strain-gauge type internal electrode pattern recently proposed will be introduced in the presentation.

Regarding drive techniques of the ceramic actuators, pulse drive and ac drive require special attention; the vibration overshoot after applying a sharp-rise step/pulse voltage onto the actuator caused a large tensile force and a long-term application of ac voltage generated considerable heat. A special pulse drive technique and/or a mechanical bias stress were required in the first case, and the heat generation had to be suppressed by changing the device design. Our analytical approach to the heat generation mechanisms will be described in the presentation. Particularly in the usage to ultrasonic motors, the antiresonance drive is highly recommended than the resonance drive, because of higher efficiency as well as lower heat generation for the same vibration level.

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# VIBRATION MODES OF TANGENTIALLY POLED PZT HOLLOW SPHERES

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Miniature size piezoelectric hollow sphere transducers have been prepared by blowing gas through a fine grained slurry of PZT-5 with a coaxial nozzle process. Binder burnout and sintering of the spheres were followed by a tangential poling of the spheres with top-to-bottom external electrode configurations.

Six different types of electrode configurations with varying electroded areas were studied and modeled. For all the configurations three principal modes of vibration were determined: an ellipsoidal mode near 230 kHz, a higher order circumferential mode at a frequency range from 300 kHz to 400 kHz, and a breathing mode near 700 kHz. Coupled modes of vibration were also determined at higher frequencies. Particularly, increasing electrode area, i.e. decreasing area of active material was found to introduce higher frequency vibrations to the admittance spectrum. Those high frequency coupled modes are: an ellipsoidal mode coupled to a thickness mode in the frequency range between 1,200 kHz to 1,350 kHz., and higher order circumferential modes coupled to thickness modes at frequencies near 2.0 MHz and 3.0 MHz. Same modes with similar frequencies were obtained from finite element analysis using the ATILA FEM code, and experimental results were shown to be consistent with the modeling study.

Hydrostatic piezoelectric charge coefficient ( $d_h$ ) of the transducers were measured and the results were found to vary between 50-160 pC/N which are several times higher than the  $d_h$  of bulk PZT. The hydrophone figure of merits ( $d_h \cdot g_h$ ) was also calculated from  $d_h$  measurements and found to vary between  $200 - 2,200 \cdot 10^{-15}$  for various types of tangentially poled spheres. These values are substantially higher than the bulk PZT figure of merit. This work was sponsored by ONR through contract # N0001492J1510.

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## NOVEL TRANSDUCER MATERIALS AND FABRICATION<sup>†</sup>

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This work presents an overview of our work in the area of high performance piezoelectric ceramics, composites, and single crystals, with an emphasis on high frequency transducers (> 20 MHz). Specific topic areas to be reviewed include the following:

- Fine Grain PZT Ceramics;
- High Frequency 1-3 Composites;
- Single Crystal Ferroelectrics with Ultra-High Electromechanical Coupling.

Novel dopant strategies and fabrication processes have resulted in  $\leq$  0.5 micron PZT-5 ceramics without hot pressing. Measurements up to 100 MHz clearly demonstrate improved performance in contrast to conventionally prepared materials.

Utilizing sol-gel technology, 1-3 composites comprised of PZT fibers on the order of 30 microns in diameter were fabricated with volume fractions  $> 30\%$ .

Relaxor ferroelectric single crystals, based on the general formula  $\text{Pb}(\text{B}_1\text{B}_2)\text{O}_3-\text{PbTiO}_3$ , have been shown to exhibit ultra-high electromechanical coupling. The reported  $K_{33}$  coefficient of  $\sim 92\%$  was recently confirmed by measurements at Hewlett Packard. Preliminary single element transducer fabrication and testing will be reported.

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# QUASISTATIC MEASUREMENTS OF THE CONVERSE AND DIRECT PIEZOELECTRIC CHARGE COEFFICIENT IN PIEZOELECTRIC CERAMICS

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This paper will describe novel experiments to measure the "quasistatic" piezoelectric charge coefficient  $d_{33}$ . The low frequency direct piezoelectric charge coefficient is found to be substantially nonlinear in stress up to 60 MPa for all the PZT types that we have studied. In addition, we have found that under short circuit operation, a large frequency dispersion exists at low frequencies (<0.1Hz). The measurement apparatus for the direct piezoelectric effect is described in detail with particular attention to improvements over previous work<sup>1</sup>. In the new experimental arrangement the current is monitored directly using a Keithly 617 electrometer insuring true short circuit conditions. The apparatus can apply both a ramp and step stress to the sample. An  $I(t) \propto k/t$  time dependence is seen after the application of a step stress. These effects are thought to result from the movement of 90° domain walls. The constant  $k$  was found to be activated with temperature. The effect of surface clamping on the sample will be discussed.

An optical lever measurement of the strain - electric field curve is also described<sup>2</sup>. A He/Ne laser is directed onto a pivoted mirror which is connected mechanically to a piezoelectric or electrostrictive material. A field is applied to the sample and the resultant strain tilts the mirror. The deflection of the laser is monitored with a linear CCD (resolution 13  $\mu\text{m}$ ) or a lateral effect detector (resolution 1.0  $\mu\text{m}$ ). Through the use of simple geometry the strain of the sample may be calculated from the laser beam deflection. The technique has the advantage over interferometric methods in that sample surfaces do not need to be mirrored and the strain range can be adjusted by changing the distance from mirror to the deflection detector. We will present our results on high field studies of piezoelectric PZT and electrostrictive PMN.

<sup>1</sup> S. Sherit, D.B. Van Nice, J.T. Graham, H. D. Wiederick, B.K. Mukherjee, Proceedings of the 8th IEEE International Symposium on the Application of Ferroelectrics, pp.167-170, Greenville, S.C., Aug, 1992

<sup>2</sup> H.D. Wiederick, S. Sherit, R.S. Stimpson, B.K. Mukherjee, Presented at the 8th International Meeting on Ferroelectricity, Nijmegen, Netherlands, July 1995, To be published in Ferroelectrics

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## MICROSCOPIC STRAIN MEASUREMENTS OF PMN-BASED ELECTROSTRICTORS

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Electrostrictors, based on solid solutions of lead magnesium niobate (PMN) with other perovskite additions, are promising candidates for use in many electromechanical devices and smart applications. The electrostrictive response of a grain in the ceramic is influenced by the surrounding grains and pores. Conventional methods, such as optical interferometry, strain gauges, and eddy-current sensors, measure only the macroscopic electrostrictive strain of the ceramic as a whole. However, X-ray diffraction can measure the response of individual crystallites to applied electric fields.

The induced lattice strain of commercial-grade PMN samples under applied static electric fields (up to 1kV/mm) was examined using X-ray diffraction. Nominal peak strain values ranged from 20 to 400 microstrain. This microscopic strain along with the polarization of the material shall be used to quantify the maximum electromechanical coupling of these materials.

Macroscopic strain measurements were conducted using strain gauge and eddy current sensor techniques. Polarization was assessed through the use of a Sawyer-Tower circuit. A digital computer with an IEEE 488 bus provided control and ease of data acquisition and manipulation. An iotech ADC 488 provided analog-to-digital conversion of the signals involved.

Samples for lattice strain measurement were electrode with a sputtered layer of Au-Pd metal approximately 80 nm thick. Samples for the macroscopic measurements, in addition to the sputtered layer, also had a fired-on silver paint coating.

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CHARACTERIZATION OF  
PMN-PT-LA (0.90 / 0.10 / 1%)  
FOR USE IN SONAR TRANSDUCERS

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For various mechanical prestresses, quasi-static room temperature measurements were made of the piezoelectric constant,  $d_{33}$ , of the relative permittivity,  $\epsilon_{33}^T/\epsilon_0$ , and of the short-circuit Young's modulus,  $Y_{33}^E$ , for PMN-PT-La (TRS Ceramics, Inc., 0.90 / 0.10 / 1%, 31 April 1995). Electric fields of up to 2 MV/m were applied to samples (2x2x10 mm) that were prestressed from 0 to 12 ksi (83 MPa). Tabulated below are the values of  $Y_{33}^E$ ,  $d_{33}$ ,  $\epsilon_{33}^T/\epsilon_0$ , and the electromechanical coupling coefficient,  $k_{33}$ , corresponding to certain mechanical prestresses, and electrical bias and drive fields. Methods for removing measurement errors because of probe resistance and because of capacitive coupling between the strain gage and the sample will be presented. [Work sponsored by the Office of Naval Research]

prestress	bias field	rms drive field	$Y_{33}^E$	$d_{33}$	$\epsilon_{33}^T/\epsilon_0$	$k_{33}$
(ksi)	(MV/m)	(MV/m rms)	(GPa)	(pm/V)		
0	0.93	0.66	86	410	15,000	0.33
4	1.0	0.62	87	420	14,000	0.36
8	1.1	0.59	88	410	12,000	0.37
12	1.3	0.55	90	400	11,000	0.38

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## DOMAIN-LIKE ORGANIZATIONS IN FERROELECTRICS AND ANTIFERROELECTRICS CONTAINING QUENCHED RANDOMNESS

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Studies of the effects of La-modification on the properties and structure have been performed across the entire lead zirconate titanate crystalline solution by dielectric spectroscopy, transmission electron microscopy, piezoelectric spectroscopy, Sawyer-Tower polarization and electrically-induced strain techniques. These studies have revealed the nature of the complex structure-property relationships in this unique family of materials for the first. These studies have shown that the effect of impurities is: (i) a common sequence of domain-like transitions between normal micron-sized domains and polar clusters with increasing disorder in the ferroelectric state, (ii) incommensuration in the polar order in the antiferroelectric state, and (iii) competing ferroelectric and antiferroelectric orderings in the region between these two states.

These results clearly indicate the presence of complicated order parameters over much of the PLZT solution and point to a means by which properties can be enhanced by control of order parameters in long-time metastable states induced by quenched disorder. The implication of these structure-property relationship studies on the design of high-performance transducers will then be discussed.